Analysis and Quantification of STE: A Novel Approach

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Abstract

The exchange of mass between the stratosphere and the troposphere has important implications for the atmospheric climate and for the life conditions. Injection of ozone-rich stratospheric air into the troposphere is recognised to significantly force the chemistry and subsequently the radiation, and can enhance episodically the ozone concentration in the boundary layer. Transport of tropospheric air (with natural and anthropogenic emissions) into the stratosphere perturbs the stratospheric chemistry and can lead to dramatic issues like the stratospheric ozone depletion.

Stratosphere-troposphere exchange (STE) has received a lot of attention during the last two decades, and numerous observational and modelling studies have been performed, which has led to an improved understanding of the subtle small-scale mechanisms which are responsible for STE. However, most attempts to quantify STE from global coverage data sets have revealed serious uncertainties.

In this thesis, the analysis and quantification of stratosphere-troposphere exchange has been performed within the Lagrangian framework. The Lagrangian diagnostic method has been developed with the goal to resolve the small-scale processes relevant to STE and to enable global coverage estimates. The use of the Lagrangian framework was motivated by two evidences. One is the difficulties encountered with Eulerian methods when attempting to quantify STE on a global scale by resolving the small-scale processes relevant to STE. The second is the intrinsic Lagrangian nature of the forcing induced by STE.

Throughout the various analyses presented in this thesis, the Lagrangian method developed and applied here has proven its potential for both the analysis and quantification of STE in the extratropics, on both hemispheric and synoptic scales. The results have revealed several striking properties of the cross-tropopause exchange flow.

First, the method has been developed and evaluated in a case study simulated with a mesoscale model and representing the evolution of a baroclinic wave, its break-up and the decay of the subsequent cut-off. Then, the meso-scale analysis of the exchange processes has led to the identification of particularly interesting mechanisms.

Secondly, the Lagrangian method has been applied to a one-year climatology (May 1995 - April 1996) using the ECMWF assimilated data. Several aspects have been analysed. The geographical structures and spatio-temporal variability of STE has been identified. Distributions of rapid transport between the stratosphere and the boundary layer (involving STE) has revealed a strong zonal asymmetry. Then the analysis of the residence time associated to the individual exchange events has allowed to establish a general residence time distribution of the flux. This intrinsic property of the exchange flux has determinant implications for the chemical and radiative forcing due to STE. The related chemical forcing
has been evaluated by use of a simple indicator.

Thirdly, the exchange mass flux has been estimated across a range of iso-PV surfaces in the upper troposphere and lowermost stratosphere. The zonal mean net flux structures have been related to the Brewer-Dobson circulation, and in addition the symmetric two-way flux has also been estimated and analysed.

Finally, ozone mass exchange has been quantified with the aid of in-situ airborne measurements, and the seasonal and geographical variability has been discussed with regard to the mass flux and ozone concentration.

The quantification of stratosphere-troposphere exchange of mass and ozone, on the hemispheric and synoptic scales have been compared with independent studies, and a good agreement has been obtained. This confirms the quantification capability of the method, while the mesoscale and climatological analyses have provided confidence in the qualitative aspects of the results.
Résumé

L'échange de masse entre la stratosphère et la troposphère a d'importantes conséquences sur le climat atmosphérique et sur les conditions de vie. Il a été reconnu que l'injection dans la troposphère d'air stratosphérique riche en ozone modifie la chimie et par conséquent la radiation. De plus, ce transport peut de temps à autre directement atteindre la couche planétaire limite. Le transport d'air troposphérique (contenant des gaz naturels et anthropogéniques) dans la stratosphère perturbe la chimie dans la stratosphère, et peut entraîner des changements importants, comme par exemple la réduction de la couche d'ozone stratosphérique.

Les échanges stratosphère troposphère (STE) ont été un centre d'intérêt durant les vingt dernières années, et un bon nombre d'études ont été menées dans des buts d'observation et de modélisation. Elles ont permis d'améliorer la compréhension des processus petite échelle qui induisent des échanges. Les études portant sur la quantification des STE à couverture globale ont révélé des incertitudes significatives.

Dans cette thèse, l'analyse et la quantification des STE a été réalisée dans un point de vue de Lagrange. La méthode diagnostique Lagrangienne a été développée dans le but de résoudre les processus petite échelle et de permettre des estimations globales. L'utilisation du point de vue de Lagrange a été motivée par deux réalités. Pour la première, il s'agit des difficultés rencontrées avec les méthodes Eulériennes lorsqu'une couverture globale est visée. La seconde est la nature intrinsèquement Lagrangienne des effets des STE.

A travers les différentes analyses présentées dans cette thèse, la méthode Lagrangienne développée et appliquée ici a démontré d'important potentiels pour l'analyse et la quantification des STE hors des tropiques aux deux échelles concernées (méso et globale). Les résultats ont révélé plusieurs propriétés importantes du flux d'échange stratosphère troposphère.

La méthode a été développée et évaluée dans le cadre d'une étude de cas sur un développement synoptique qui représente l'évolution d'une onde baroclinique, sa rupture et la décroissance du "cut-off" postérieur. Ensuite, l'analyse méso-échelle des processus d'échange a conduit à l'identification de mécanismes particulièrement intéressants.

La méthode a ensuite été appliquée dans le cadre d'une climatologie couvrant une année (mai 1995 à avril 1996) basée sur les données d'assimilation de l'ECMWF. Différents aspects ont été analysés. La structure géographique et la variabilité spatio-temporelle des STE ont été identifiées. L'estimation du transport rapide entre la stratosphère et la couche planétaire limite ont montré de fortes asymétries zonales. Ensuite, l'analyse des temps de résidence associés aux événements d'échange ont permis d'établir une distribution de temps de résidence du flux. Cette propriété intrinsèque du flux d'échange a des conséquences déterminantes pour la chimie et la radiation induite par les STE. La perturbation de la chimie a été estimée avec un indicateur simple.
De plus, le flux d'échange a été estimé à travers plusieurs niveaux de constant tourbillon potentiel (PV) dans la haute troposphère et la basse stratosphère. Les structures du flux net, moyenné zonalement, ont été interprétées en terme de circulation générale de Brewer-Dobson. Le flux d'échange complémentaire “two-way” symétrique a aussi été estimé et analysé.

Finalement, la masse d'ozone échangée a été quantifiée à partir de mesures prises d’avion, et les variabilités saisonnière et géographique ont été discutée relativement au flux de masse et à la concentration d’ozone.

Les quantifications d’échange stratosphère troposphère de masse et d’ozone, à l’échelle hémisphérique et synoptique ont été comparées à des études indépendantes avec succès. L’accord entre ces estimations confirme la capacité quantitative de la méthode, alors que les analyses qualitatives faites aux échelles hémisphérique et méso ont donné confiance dans les aspects qualitatifs des résultats.
Chapter 1

Introduction

1.1 Preamble

The troposphere and the stratosphere possess very different properties and their coupling is determinant for the dynamics, chemistry and radiative budgets of the atmosphere. The troposphere is characterised by a variety of gases (among others water vapour, CO$_2$, organic compounds and other anthropogenic gases) and a low static stability allowing the vertical mixing. The stratosphere is much more stratified and is composed of a relative small variety of different chemical constituents, with very low water vapour levels and high ozone concentrations. The quasi-material separation of these two atmospheric layers is insured by the presence of a minimum in the temperature lapse rate. By convention of the World Meteorological Organization (WMO 1985) the tropopause is defined as the lowest level at which the temperature lapse rate decreases to 2 K km$^{-1}$ or less and the lapse rate averaged between this level and any level within the next 2 km does not exceed 2 K km$^{-1}$.

The different chemical nature of the troposphere and stratosphere, which reflects their isolation from each other, indicates that exchange of mass between them may significantly affect the chemistry and in turn the radiative flux balance. Early observations of inflow of stratospheric mass into the extratropical troposphere have been reported by Danielsen (1968). They measured radioactivity from an aircraft in the region of an upper-level front (e.g. Keyser and Shapiro 1986) and showed that a substantial amount of the radionuclide strontium-90, which was injected in the stratosphere during bomb tests, entered the troposphere.

The Definitions of the Tropopause

The identification and quantification of the exchanged mass requires the unambiguous localisation of the tropopause, also in jet stream regions where the thermal definition of the tropopause breaks down. To enable such analyses and to better rationalise the quasi-material behaviour of the tropopause, a dynamical definition of the tropopause is often applied instead of the thermal one. The dynamical tropopause is defined by a surface of constant potential vorticity (PV) (cf. Hoskins et al. 1985 for a review) in the extratropics and by an isentropic surface in the tropics. Usually, the isentropic surface 380 K and the surface of constant potential vorticity 2PVU, where PVU denotes the standard potential vorticity unit.
Figure 1.1: Contours of the annual and zonal mean potential vorticity (2PVU, bold line) and potential temperature (thin lines) fields, as calculated for the period May 1995 to April 1996. Potential temperature contours are shown every 5 K between 280 - 370 K and every 10 K between 370 - 480 K, and the 380 K is highlighted.

(1 PVU = 10^{-6} \text{ m}^2\text{s}^{-1}\text{Kkg}^{-1}), are used due to their closeness with the thermal tropopause (e.g. Holton et al. 1995). The dynamical properties of the potential vorticity allow furthermore an improved dynamical interpretation of the exchange processes. This will be discussed in Chapter 2.

Figure 1.1 represents the zonal mean 2PVU iso-surface and isentropes. The tropopause is steepest in the subtropical region (2PVU iso-surface), and is low and warm in the extratropics (2PVU iso-surface) and higher and cooler in the tropics (380 K isentrope). The region enclosed between the 2PVU and the 380 K iso-surfaces in the extratropics is called the “lowermost stratosphere”, and the region above the 380 K isentrope is called the “overworld”, according to Holton et al. (1995). Note that only the isentropic surfaces between about 300 K and 380 K cross the tropopause (Hoskins 1991).

The Processes Responsible for STE

Lots of studies have attempted during the past two decades to identify the physical processes responsible for stratosphere-troposphere exchange (STE).\footnote{Aspects concerning the diagnosis of STE are treated in Chapter 2.} They can be classified in four groups regarding the target scale and the device: (i) the in-situ or remote observation of a stratospheric or tropospheric tracer principally via aircraft, ozonesonde and lidar; (ii) the analysis of mesoscale model simulations; (iii) the analysis of global coverage assimilated meteorological data which include the tropopause region; and (iv) the analysis of satellite

\footnote{The data assimilation-initialisation-forecast cycle incorporates observational data from various operational sources (surface stations, radiosondes, balloon and satellite soundings, commercial aircraft and drifting buoys) and produces consistent meteorological data sets.}
data. In-situ measurements and mesoscale models provide insight on the small-scale processes but have a limited coverage. They have been extensively used to study synoptic- and meso-scale aspects of STE. On the other hand assimilated meteorological and satellite data have been used mainly to study large-scale mixing aspects relevant to STE without emphasis on the small-scale features.\(^3\)

In the present study the focus is placed on the stratosphere-troposphere exchange in the extratropical and subtropical region. To give an overview of the physics and dynamics of exchange, a brief review of existent studies is provided hereafter for the extratropical and subtropical region. A review of the exchange processes occuring in the tropics can be found in Holton et al. (1995).

In the extratropics, the injection of stratospheric air into the troposphere has been detected almost exclusively in regions of upper-level troughs and cut-off lows associated with an enhanced baroclinicity (e.g. Danielsen 1980, Shapiro 1980, Ancellet et al. 1991, Langford et al. 1996, Eisele et al. 1999). The life-cycle of these baroclinic waves has been shown to be associated with the production of subsequent folding and filamentation structures (e.g. Bush and Peltier 1994, Bithell et al. 1999). The physical processes responsible for stratosphere-troposphere exchange have been analysed in numerous studies and a variety of mechanisms have been identified, while their relative importance varies significantly from case to case. The presence of clouds in the upper-troposphere (often induced on the warm side of an upper-level trough) and the associated diabatic effects lead generally to a significant inflow of stratospheric air into the troposphere (e.g. Lamarque and Hess 1994, Wirth 1995b). Turbulence in the region of the jet stream occurring in absence of clouds has been identified by Shapiro (1976) (clear air turbulence) and its significance for STE has been demonstrated in Shapiro (1980). The formation of folds and their irreversible mixing is also recognised to account for a significant part of STE (e.g. Price and Vaughan 1993, Hartjenstein 2000). The importance of these tropopause structures for STE has led to attempts of quantifying globally STE using climatologies of the frequency of occurrence of cut-off lows and tropopause folds (Price and Vaughan 1992, Beekmann et al. 1997, Elbern et al. 1998).

The presence of filaments of stratospheric air in the troposphere in the region of the tropopause has been first detected from satellite data of water vapour and isentropic PV fields calculated from ECMWF assimilated data (Appenzeller and Davies 1992), and from the water vapour field of ECMWF assimilated data (Newell et al. 1992). These small-scale filamentation structures are identifiable on sufficiently high resolution isentropic PV charts and on water vapour satellite imagery, and evolve from upper-level disturbances like troughs (or "streamers" on an isentropic PV chart) and cut-off lows. Due to their small-scale nature, they are rapidly and irreversibly mixed in the troposphere (e.g. Forster and Wirth 2000, Eser et al. 1999), and are therefore relevant for STE (Appenzeller et al. 1996a, Wirth 1996). The quantification of the total surface of filaments on the global scale provides a measure of mixing of stratospheric air into the troposphere.\(^4\)

Comparatively few studies have analysed the physical processes involved in troposphere-

\(^3\)Note that methods attempting to diagnose STE with a large-scale coverage and resolving the synoptic-scale processes still have large uncertainties (cf. Chapter 2).

\(^4\)The method called "Contour Advection Technique" is generally used to generate these PV filaments from relative low resolution data by the advection on an isentropic surface of the PV-contours.
to-stratosphere exchange in the extratropics. Radiative effects in anti-cyclones, in particular those related to the water vapour profile, are known to induce exchange (e.g. Hoskins et al. 1985, Zierl and Wirth 1997). Additionally, strong convective complex which overshoot the tropopause are known to inject moist tropospheric air into the stratosphere at the anvil outflow, a process which is likely to play an important role in the stratospheric water vapour budget (e.g. Poulida et al. 1996, Stenchikov et al. 1996).

Gravity waves have recently received attention, thanks to the development of high resolution non-hydrostatic models. Breaking of gravity waves (for instance topographically excited) in the region of the tropopause has a significant potential for inducing STE (Lamarque et al. 1996, Moustaoui et al. 1999).

Stratosphere-troposphere exchange in the subtropics is not associated with deep stratospheric intrusions but is likely to take place quasi-isentropically across the particularly steep tropopause found in this region. Despite the strong gradient of PV which acts to inhibit the cross-tropopause transport, troposphere-to-stratosphere exchange has been detected (e.g. Dessler 1996, Folkins and Appenzeller 1996), and mainly attributed to the poleward advection of upper-tropospheric air during Rossby-wave breaking events (e.g. Peters and Waugh 1996, Vaughan and Timmis 1998, O'Connor et al. 1999), or related to the Asian summer monsoon (Dethof et al. 1999).

The Large-Scale Circulation and STE
The small-scale processes induce a net mass flux across the tropopause which in turn can be regarded as part of the general mass circulation in the atmosphere. The mean meridional tracer circulation was proposed by Brewer (1949) and Dobson (1956) in the form of a single mean meridional cell in each hemisphere with a global rising motion in the tropics, a poleward drift in the stratosphere, and by continuity of mass a return flow into the troposphere in the extratropics (see broad arrows in Fig. 1.2). The general single cell shape of the Brewer-Dobson tracer circulation has been confirmed later in various studies (see Chapter 2). Furthermore, it has been shown by Haynes et al. (1991) that the general mass circulation can be regarded as being driven by an “extratropical pump” in the stratosphere itself generated by the breaking of large-scale Rossby waves in the extratropical stratosphere (see Chapter 2). From this large-scale viewpoint the stratosphere-troposphere exchange is regarded as a net transport by eddy motions in the tropopause region (see wiggly arrows in Fig. 1.2) and is related to the general circulation via the hemispheric and annual mass budget in the lowermost stratosphere.

However, the large-scale (and also regional scale) effects of stratosphere-troposphere exchange are intimately related to the chemical constituents which are transported. Thus, the separate knowledge of the stratosphere-to-troposphere and the troposphere-to-stratosphere exchange fluxes is fundamental for issues related to the global chemistry and global radiative budget. Such a separate evaluation of up- and downward STE requires to resolve the small-scale processes which are responsible for STE.

Recent developments of general circulation models (GCM) coupled with chemical models have shown that the quantitative evaluation of separate up- and downward STE fluxes with the (often) rough physics and dynamics of the model is a challenging task but results are generally encouraging (Mote et al. 1994, Cox et al. 1997, Rind et al. 1999). Such global
1.2 AIMS OF THIS STUDY

Figure 1.2: Large-scale circulation and stratosphere-troposphere exchange. The tropopause is shown by the thick line and the isentropes by the thin lines. The heavily shaded region represents the lowermost stratosphere. Light shading in the overworld denotes wave-induced forcing (the "extratropical pump"). The wiggly double headed arrows denote meridional transport by eddy motions. Broad arrows show transport by the global-scale circulation. From Holton et al. (1995).

coupled models, which permit to gain insight in the dominant forcings of climate, have demonstrated that STE plays a key role in the tropospheric chemistry and radiative budgets: ozone of stratospheric origin is evaluated to about 40% of the total ozone found in the troposphere (Roelofs and Lelieveld 1997).

1.2 Aims of this Study

Thus, stratosphere-troposphere exchange has strong effects in the atmosphere which are important for the global climate and also for the life conditions in the boundary layer. The massive injection of ozone into the troposphere forces the global tropospheric chemistry and induces large-scale radiative effects. On the regional scale, the episodic deep intrusion of ozone can dramatically increase the ozone levels in the low troposphere, and combined with the anthropogenic emissions, this can affect locally the life conditions (e.g. Randriambelo et al. 1999). The injection of tropospheric air into the stratosphere has numerous effects, one of the most important being the stratospheric ozone depletion attributed to the presence of chlorofluorocarbons (CFCs)
(cf. Solomon 1999 for a review). Moreover, significant radiative effects are induced in the relative cold and dry lowermost stratosphere by the injection of moisture and aerosols from the troposphere. And chemical perturbations of the lowermost stratosphere have been detected in relation to the exchange of various anthropogenic species (Lelieveld et al. 1997).

These important effects of STE and their consequences on the climate and its prediction are all relevant motivations which have driven for the two past decades the attention of numerous scientists on this specific aspect of the atmosphere. The physical processes responsible for STE have been extensively studied and are now generally considered as roughly understood. The large-scale quantification of STE has become an important and challenging goal. Attempts of estimating STE by resolving the small-scale processes at the tropopause level with a global coverage are still subject to large uncertainties (Chapter 2).

In this study, the analysis and quantification of STE is approached within the Lagrangian framework, as opposed to most past studies which are cast in the Eulerian framework.

One thrust of this study is the development of a Lagrangian diagnostic method for STE which enables realistic hemispheric-scale estimates based upon the synoptic-scale physics and dynamics. The other main objective is to draw the fundamental Lagrangian properties of STE.

The various aspects of STE that are analysed in this study are dressed in Chapters 3 to 9, prior to which, in Chapter 2, the details of the existing methods attempting to diagnose STE are discussed. First, the development of the method and the related methodological analyses are discussed in Chapter 3. In Chapter 4 a detailed analysis is provided of STE resulting from the application of the diagnostic method to a synoptic-scale case study. This case study both evidences subtle exchange mechanisms and allows the validation of the method on the meso-scale. Then, several climatological aspects of STE are treated in Chapters 5 to 9. In Chapter 5 the method is applied to derive a one-year climatology on the northern hemisphere. The basic estimates are analysed and a comparison with an independent study permits the validation of the method on the large-scale. Chapter 6 reveals an important Lagrangian property of STE: the residence time distribution. In Chapter 7 STE is generalised to the transport across PV iso-surfaces in the upper-troposphere lower stratosphere region. Then, an estimation of the ozone exchange mass flux across the tropopause is presented and characterised in Chapter 8. And finally, an attempt of estimating the actual chemical forcing due to STE based upon the associated residence time distribution is proposed in Chapter 9.
Chapter 2
Diagnosis of Stratosphere - Troposphere Exchange

Before discussing the approach adopted in this thesis, it is appropriate to review the existent methods used to estimate STE. Apart from Lagrangian methodologies based upon the analysis of trajectory, there are two other kind of methods: those which operate within the zonal mean theoretical framework and provide zonal mean climatologies, and those based on local Eulerian formulation and seek to provide synoptic scale estimates of the STE flux. These approaches can be used for either a synoptic scale or a large scale quantitative comparison, and they can provide insight on the physics of STE. The goal here is to discuss the diagnostic methods that have been used in the past to estimate STE and to understand their limitations. The last section will then place the Lagrangian method within this broad context and discuss earlier work conducted within this framework.

2.1 Basic Notions

The basic governing equations that describe atmospheric flow are the so-called primitive equations comprising the momentum equations (2.1), the mass continuity (2.2) and the thermodynamic equation (2.3):

\[
\frac{Du}{Dt} + 2\Omega \times u = -\frac{1}{\rho} \nabla p - \nabla \Phi + F
\] (2.1)

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0
\] (2.2)

\[
\frac{D\theta}{Dt} = \hat{\theta},
\] (2.3)

where \(\frac{D}{Dt}\) denotes the material derivative following a fluid parcel, \(u\) is the 3-dimensional flow velocity, \(\rho\) the mass density, \(\Omega\) the angular velocity of the earth, \(\theta\) the potential temperature, \(p\) the pressure, \(\Phi\) represents the geopotential \(gz\) (or the potential of other conservative body forces) and the non-conservative terms \(F\) and \(\hat{\theta}\) are the friction and the diabatic heating sources, respectively.
Evaluating the scalar product of the curl of (2.1) with $\nabla \theta$ leads to the following fundamental equation:

$$\frac{DQ}{Dt} = \frac{1}{\rho} \{ \omega \cdot \nabla \theta + \nabla \times F \cdot \nabla \theta \}$$  \hspace{1cm} (2.4)

where $Q$ is the Ertel's potential vorticity (PV), defined by

$$Q = \frac{1}{\rho} \omega \cdot \nabla \theta \hspace{1cm} (2.5)$$

and $\omega = 2\Omega + \nabla \times u$ is the absolute vorticity. As can be inferred from (2.4), the potential vorticity $Q$ of a parcel is materially conserved under adiabatic frictionless conditions ($F = 0$ and $\theta = 0$). This fundamental property renders PV useful for studying atmospheric flow dynamics on a wide variety of scales. Although potential vorticity and its conservation property have been early recognised (Rossby 1940, Ertel 1942), its wide use as a fundamental dynamical variable was stimulated later by the work of Hoskins et al. (1985). They discussed the key properties of $Q$ such as the invertibility principle, and presented examples of the analysis of typical synoptic situations using the PV framework.

The large-scale vertical structure of potential vorticity is dominated by the profile of $\rho^{-1} \partial \theta / \partial z$, with low values in the troposphere of the order of magnitude of 1PVU (1PVU = $10^{-6}$ m$^2$ K s$^{-1}$ kg$^{-1}$), and rapidly increasing values above the tropopause. This overall vertical structure together with its quasi-conserved nature makes PV a favourable candidate to define the tropopause (as an alternative to the thermal definition, see Chapter 1). Indeed, a dynamical tropopause defined by an iso-PV surface has the following advantages, compared to the thermal definition: it gives a clear physical meaning to the nature of exchange processes across the tropopause via (2.4), and it results in a continuous tropopause surface, also in upper level frontal zones. These two properties of the dynamical tropopause are crucial to establish a proper definition of stratosphere-troposphere exchange events, unambiguous at mesoscale. Due to the large PV gradients at the tropopause level, different values have been chosen for the tropopause definition. However, STE fluxes estimated at different iso-PV surfaces can possibly lead to different values, depending on the amplitude of the non-conservative processes. In most studies aiming to estimate STE at the synoptic scale, a dynamical tropopause has been chosen with a threshold value varying between 1.6 and 4PVU.

The conservation property of Ertel's PV can be generalised to a class of quantities defined by replacing the potential temperature in $Q$ by a function $f(\theta)$ of it:

$$Q = \frac{1}{\rho} \omega \cdot \nabla f(\theta). \hspace{1cm} (2.6)$$

As suggested by Haynes and McIntyre (1990), it is likely that a different choice of PV could still better mark on average the tropopause. Estimates of STE using alternative expressions for PV than the Ertel's one would be of course possible, and in particular with the method proposed in this study, and an analysis of the dependency of STE with the function $f(\theta)$ in the potential vorticity expression would be interesting for fundamental dynamical purposes. However, this question is beyond the scope of our study and the Ertel's potential vorticity is assumed to reasonably represent the tropopause in the following.
Beside tropopause-marking issues, the conservation property of PV is a particularly interesting tool for the analysis of atmospheric flow motion. Specifically, the material conservation property of PV implicates for a flow motion which is dominated by its adiabatic and frictionless component to be constrained to remain on an iso-PV surface. Mapping the intersection contours of iso-PV surfaces with isentropes, for instance by representing PV in isentropic coordinates, yields consequently a direct and simple view of the atmospheric flow. The isentropic PV (IPV) has been widely used for synoptic- and global-scale flow analyses (for reviews, see Hoskins et al. 1985, Hoskins 1991). For the stratosphere-troposphere exchange issue, the IPV framework allows to gain interesting insights in the exchange processes, both on the large- and the synoptic-scales. On the large-scale, non-conservative processes implying material changes in PV are assumed to be predominantly associated either to the diabatic circulation (see for instance Yang and Tung 1996), or to (quasi-) isentropic mixing (see for instance Chen 1995). Isentropic mixing in turn can be seen in an IPV perspective as an irreversible deformation and subsequent mixing of PV contours (Appenzeller et al. 1996a). On the synoptic-scale, the IPV view of the evolution of the tropopause offers a useful information on the presence of non-conservative sources, via the irreversible deformation of PV contours.

An alternative formulation of (2.4) based upon the PV substance (PVS) defined as $\rho Q$, was set out by Haynes and McIntyre (1987; 1990). They showed (2.4) can be rewritten in the following conservative form without restrictions concerning the nature of the non-conservative terms $F$ and $\dot{\theta}$:

$$\frac{\partial \rho Q}{\partial t} + \nabla \cdot J^{PVS} = 0 \tag{2.7}$$

where the flux of PVS $J^{PVS}$ is given by:

$$J^{PVS} = J^{ADV} + J^{NA} \tag{2.8}$$
$$J^{ADV} = \rho Q u \tag{2.9}$$
$$J^{NA} = -\omega \theta - F \wedge \nabla \theta \equiv \rho Q u^{NA} \tag{2.10}$$

The arbitrariness of the PVS flux formulation which comes from the introduction of the non-conservative terms inside the divergence in (2.7) has been analysed by Bretherton and Schär (1993). They found that the PVS flux formulated originally by Haynes and McIntyre (1987) is the unique choice that can be written as the sum of a purely advective ($J^{ADV}$) and a non-advective ($J^{NA}$) flux with the latter depending linearly on local heating rate and frictional forces. It is therefore particularly appropriate for the physical interpretation.

An additional property of the PVS has been emphasised by Haynes and McIntyre (1987) which is the impermeability of isentropic surfaces to the PVS. With these characteristics, PVS can be viewed as a pseudo-material substance which is advected with the isentropic component of the air motion. The value of PV corresponds to the mixing ratio of PVS. Thus, for example, the advection of a positive PV anomaly can be viewed as the advection of an air mass with high concentration of PVS, and the erosion of a cut-off low by non-conservative processes can be interpreted as a cross-isentropic motion of air into the concerned isentropic layer which acts to dilute the PVS in the layer, like a “sponge”. Conversely, the decay of a negative PV anomaly can be viewed as a cross-isentropic transport of air out of the concerned
isentropic layer which acts to concentrate the PVS, like an "antisponge". Applying this mechanism in a broader context, the global diabatic circulation is brought in a new light: the general tendency for positive PV anomalies to be advected equatorward and negative anomalies to be advected poleward, both decaying diabatically as they travel, implies globally that a given isentropic layer takes up mass in low latitudes and expels it in high latitudes. And the vertical mass flux across a given isentropic surface resulting from the cross-isentropic fluxes involved in PVS dilution / concentration above is the diabatic circulation. The diabatic circulation can consequently be thought as being driven by the equatorward eddy flux of PV. This idea, further discussed in the next section, is the downward control principle.

2.2 Methods Cast in the Zonal Mean Framework

2.2.1 The Transformed Eulerian Mean Equations

Zonally averaging the primitive equations leads to a system which describes the mean atmospheric motion in a meridional plane. This particular form of the atmospheric flow equations can be used to estimate exchange fluxes across some given surface, and some global scale properties of exchange and the associated processes can be inferred. Caution is required on how to built the zonal average of the flow equations because the mean meridional motion of a tracer is obviously not directly given by the Eulerian zonal mean of the winds, as defined by:

\[ \bar{u}(\phi, z, t) = \frac{1}{2\pi} \int_0^{2\pi} u(\lambda, \phi, z, t) d\lambda \] (2.11)

\[ u'(\lambda, \phi, z, t) = u - \bar{u}, \] (2.12)

where \( \lambda \) and \( \phi \) are longitude and latitude, respectively, and \( z \) denotes height. The zonal mean of the material transport of a species with mixing ratio \( \chi \) and source \( S \) takes in fact the form:

\[ \frac{\partial \bar{\chi}}{\partial t} + \bar{\theta} \frac{\partial \bar{\chi}}{\partial \theta} + \bar{w} \frac{\partial \bar{\chi}}{\partial z} = \bar{S} - \rho_0^{-1} \nabla \cdot (\rho_0 w \bar{\chi}') \] (2.14)

The left hand side of this equation is the material change of \( \chi \) following the Eulerian zonally averaged circulation, and consequently, the right hand side describes its sources. Thus, beside the natural tracer source \( \bar{S} \), there is an additional source, the divergence of the zonal eddy tracer flux, which is purely due to the zonal motion of the tracer and is not related to a real source of \( \chi \).

This problem is removed to some extent under some assumptions (linear, steady and adiabatic atmospheric waves) by using instead the transformed Eulerian mean (TEM) (see Andrews et al. 1987). The residual mean velocities in log pressure coordinate are defined as

\[ \bar{v}^* = \bar{v} - \frac{1}{\rho_0} \frac{\partial}{\partial z} \left( \rho_0 \bar{u}' / \bar{\theta}' \right) \] (2.15)

\[ \bar{w}^* = \bar{w} + \frac{1}{a \cos \phi} \frac{\partial}{\partial \phi} \left( \cos \phi \bar{u}' / \bar{\theta}' \right) \] (2.16)
and the zonal momentum, the thermodynamic and the continuity equations (2.1, 2.2, 2.3) become (in spherical coordinates):

\[
\frac{\partial \bar{u}}{\partial t} + \bar{v}^* \left[ \frac{1}{a \cos \phi} \frac{\partial}{\partial \phi} (\bar{u} \cos \phi) - f \right] + \bar{w} \frac{\partial \bar{u}}{\partial z} = \frac{1}{\rho_0 a \cos \phi} \nabla \cdot \mathbf{P} + \bar{X} = \mathcal{F} \tag{2.17}
\]

\[
\frac{\partial \bar{\theta}}{\partial t} + \frac{\bar{v}^*}{a} \frac{\partial \bar{\theta}}{\partial \phi} + \bar{w} \frac{\partial \bar{\theta}}{\partial z} = \bar{\theta} - \frac{1}{\rho_0} \frac{\partial}{\partial z} \left[ \rho_0 \left( \bar{v}^* \frac{\partial \bar{\theta}}{\partial \phi} + \bar{w}' \bar{\theta}' \right) \right] \tag{2.18}
\]

\[
\frac{1}{a \cos \phi} \frac{\partial}{\partial \phi} (\bar{v}^* \cos \phi) + \frac{1}{\rho_0} \frac{\partial}{\partial z} \left( \rho_0 \bar{w}^* \right) = 0 \tag{2.19}
\]

The source of momentum \( \mathcal{F} \) is composed of two terms, \( \nabla \cdot \mathbf{P} \) the divergence of the Eliassen Palm flux (EP flux) (see Andrews et al. 1987) and \( \bar{X} \) the zonal mean zonal component of \( \mathbf{F} \) (see 2.1), comprising unresolved effects such as gravity wave drag, friction, and other non-conservative mechanical processes. The source terms of potential temperature comprise the zonal mean heating rate \( \bar{\theta} \) and an additional term due to eddy fluxes of heat.

### 2.2.2 Flux Estimation Based on the Diabatic Heating \( \bar{\theta} \)

A first approach to analyse this system of equations is to combine the thermodynamic equation with the continuity equation. The flux divergence term in the thermodynamic equation (2.18) is small under quasi-geostrophic scaling (i.e. for large-scale systems) and if it is ignored, these two equations can be solved for the residual velocities provided an accurate estimate of \( \bar{\theta} \) is available (Rosenlof 1995). A correction is generally made to the residual velocities in order to ensure that the net vertical mass flux across a pressure surface is zero.

Estimation of the diabatic heating rates \( \bar{\theta} \) is a delicate issue and is usually provided by two-dimensional sophisticated radiative transfer codes using global temperature fields and concentrations of radiatively active constituents such as \( \text{O}_3 \), \( \text{H}_2\text{O} \), \( \text{CH}_4 \), \( \text{NO}_x \), \( \text{CO}_2 \), as well as of stratospheric aerosols (see Rosenlof 1995, Appenzeller et al. 1996b, Eluszkiewicz et al. 1996, Eluszkiewicz et al. 1997). Although this method provides fairly stable results in the northern hemisphere middle atmosphere (Eluszkiewicz et al. 1997), the quality is strongly dependent on the accuracy of the radiative transfer model because the net radiative heating is obtained by subtracting the almost-equal solar heating and thermal cooling rates at each stratospheric level. The uncertainty becomes particularly large in the lower stratosphere where these two terms differ by less than 20% (Eluszkiewicz et al. 1997). This cancellation problem together with the role of cloud-diabatic heating which become non-negligible in the lowermost stratosphere renders the method uncertain in this region.

The resulting estimates of the residual meridional circulation led to the general picture of the mean transport in the middle atmosphere (which is qualitatively in close agreement with the early representation of the mass circulation of Brewer (1949) and Dobson (1956)): a large scale upwelling in the tropics and downwelling in the extratropics. Figure 2.1 shows the monthly residual circulation streamfunction above 100hPa calculated by Eluszkiewicz et al. (1997) for the period January 1992 to May 1993. At the solstices, the circulation is dominated by one cell centred on the winter hemisphere, while it is composed of two more
or less symmetric cells at the equinoxes. Some indicative values of mass fluxes are given in Table 2.1.

2.2.3 Flux Estimation Based on the Momentum Forcing $\mathcal{F}$

The second method is based upon the *downward control* principle proposed by Haynes and McIntyre (1987) and McIntyre (1987), and analysed in details by Haynes et al. (1991). Here, the zonal momentum (2.17) and the continuity equation (2.19) are solved for $(\tilde{v}^*, \tilde{w}^*)$, assuming that the momentum forcing $\mathcal{F}$ can be estimated from observed perturbation winds. The calculation does not depend upon an estimate for the heating, and its accuracy holds in the realization of its assumptions as well as the quality of the observed winds used to compute the forcing. In steady state conditions the downward control principle links the extratropical mass flow across an isentropic surface to the momentum forcing distribution (the *extratropical pump*) above this surface. In this formulation, the frequency of tropopause folding below the forcing level (which is assumed to be in the stratosphere) does not establish the mass flux across an isentrope in the lower stratosphere. The principle is applicable in regions where angular momentum contours are vertical and therefore not appropriate in the tropics. The robustness of the principle has been examined in cases where the momentum forcing is time-dependent (Haynes et al. 1991) and it was found that the principle is remarkably robust and can be applied to the real atmosphere on the time scale of months for large horizontal scale. When a fluctuating force is applied in a shallow layer, the control is at least 84% downward. On the other hand, the penetration through a 7 km height of a large scale circulation after a sudden change in the forcing above takes about 6 days, and the adjustment time for the atmosphere to approach the steady state is about 20 days. However, the adjustment time has been shown to increase as the horizontal scale of the forcing decreases, and for a forcing reduced from the initial 30° latitude to a horizontal scale of about 10° latitude, the model takes about 50 days to reach steady state conditions. The dominant forcing for the midlatitude stratospheric circulation is the wave drag associated with the breaking of large-scale Rossby waves in the winter hemisphere. An illuminating view of the momentum forcing term can be gained from rewriting the EP flux divergence under quasi-geostrophic scaling:

$$\nabla q^e = \mathbf{a}_0^{-1} \nabla \cdot \mathbf{p}.$$  \hspace{1cm} (2.20)

The momentum forcing can thus be interpreted as the northward eddy flux of (quasi-geostrophic) potential vorticity $\nabla q^e$, the latter being a statistical measure for the equatorward (poleward) travelling of positive (negative) PV anomalies as can be observed on isentropic potential vorticity (IPV) maps in the stratosphere. The dominant contribution to the momentum source is confined to the midlatitude winter hemisphere where the incidence of wave breaking is high (McIntyre and Palmer 1983, Juckes and McIntyre 1987). This region of the stratosphere surrounding the main stratospheric vortex has been termed *stratospheric surf zone* by McIntyre and Palmer (1983) by analogy with the surf zone of an ocean beach. This surf zone has two well defined edges evidenced by the strong PV gradients at high latitude (the polar vortex edge) and at low latitudes near 10°-20° (the subtropical PV barrier) (Randel et al. 1993). Thus, on the equatorial side of the extratropical pump, the downward control principle, in its steady state, would predict an upwelling in latitude bands much closer to
Figure 2.1: Latitude-height sections of the mass-weighted streamfunction (in units of $10^9$ kg/s). The contour interval is 0.2 and 1.0 for absolute values lower and greater than 2.0, respectively. The zero line is thicker and shading denotes areas of counterclockwise flow. (From Eluszkiewicz et al. 1997.)
Figure 2.2: Schematic of the steady meridional circulation produced by midlatitude wave drag. Wave drag is confined to the winter midlatitude surf zone, denoted by the shading. Arrows depict the circulation, and vertical thin lines represent iso-lines of the angular momentum. See text for discussion of cases (a) and (b). (From Plumb and Eluszkiewicz 1999.)

the momentum source zone (Fig. 2.2 a) than has been diagnosed from diabatic calculations (Figs. 2.1 and 2.2 b). Indeed Plumb and Eluszkiewicz (1999) suggest that viscous contributions to $\mathcal{F}$ can act to redistribute the circulation in the tropics and that stratospheric (and perhaps tropospheric) heating in the tropics could even make a significant contribution to the net upwelling. Rosenlof and Holton (1993) estimated the residual circulation in a GCM (the NCAR CCM2 model) using the downward control method and compared results with estimations of $\mathbf{v}^*$ using (2.15), (a more direct computation due to the availability of relevant model parameters). Good agreement was found for the solstice seasons, while for the equinox seasons the steady state assumption was found to be violated due to the net change in the zonal wind through the season. In addition, they suggested that the gravity wave drag contribution to the total forcing is significant in particular for the latitudinal distribution of downward fluxes in the extratropics and therefore a parameterisation of this sub-grid forcing should be included in the computation. Table 2.1 provides a summary of estimates of extratropical downward fluxes across the 100hPa surface using the downward control principle from Rosenlof and Holton (1993) and Holton (1990).

## 2.2.4 Link to the Cross-Tropopause Mass Flux

Estimates of the residual circulation permits the quantification of the net mass flux across an isobaric or an isentropic surface lying within the stratosphere. However, quantification of the flux across a realistic tropopause demands a further step. Indeed, although the net mass flux across the tropopause is expected to equalise the net mass flux across the 380 K isentropic surface on long time scales, this is not necessarily true for time scales of months or seasons. Appenzeller et al. (1996b) examined the seasonal variation of the mass of the
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Table 2.1: Estimates from the literature of global scale extratropical downward mass flux across various surfaces (exch surf) in 10^8 kg/s. The values in bracket corresponds to the minimum latitude of the integration (which also represents the latitude where the flux changes direction). UKMO: United Kingdom Meteorological Center assimilated data, NMC: National Meteorological Center of USA stratospheric analyses, MLS: data from the Microwave Limb Sounder onboard the Upper Atmosphere Research Satellite (UARS), CLAES: data from the Cryogenic Limb Array Etalon Spectrometer onboard the UARS, Oort: atlas of global atmospheric circulation statistics for 1958-1973 (Oort’s 1983).

lower stratosphere and compared it to the variation of the mass flux at 380 K calculated from a radiative transfer code. The result is summarised in Fig. 2.3, where $F_{380K}$ is the downward flux at 380 K, $F_{out}$ is the final downward flux across the tropopause and $F_{90Sr}$ is a northern hemisphere estimate based on representative measurements of $^{90}$Sr mixing ratios. The seasonal shift between the two fluxes is striking: whilst the flux at 380 K reaches its maximum in winter, the flux at the tropopause is maximum in spring. Thus, the flux at 380 K cannot be used as a proxy of the flux at the tropopause level on the seasonal time scale.

### 2.2.5 Further Remarks

Finally, the global comparison of the estimates of extratropical downward flux found in the literature (Table 2.1) show values generally larger at 100hPa than at 70hPa and a robust seasonal cycle, with a maximum in winter and a minimum in summer. However, differences between the estimates of flux across equal surfaces can be significant and thereby confirm the uncertainties discussed above. In particular, large disagreement is found for the summer
Figure 2.3: Annual cycle of the net downward mass flux across the northern hemisphere extratropical tropopause ($F_{\text{out}}$, thick plain curve), the 380 K isentropic surface ($F_{380}$, thin dashed curve), and mass flux across the tropopause estimated from $^{90}$Sr measurements ($F_{90Sr}$, dotted line). Shown are the two years 1992 and 1993. Negative values denote downward mass flux. (From Appenzeller et al. 1996.)

at 100hPa, where a significant contribution to the diabatic heating is expected from clouds and convection.

2.3 Eulerian Methods Based on a Local Formulation of STE

Tropospheric and stratospheric chemistry is likely to be susceptible to the zonal, latitudinal and seasonal variability of STE and therefore there is a need for methods which are able to provide geographically resolved estimates of STE.

2.3.1 The Wei Formulation

The most commonly used Eulerian formulation of STE was developed by Wei (1987). The formulation (hereafter referred to as the Wei-formulation) is based on a generalised coordinate system $(x, y, \eta, t)$, where $\eta$ is a global vertical coordinate. Furthermore, a functional relationship between the potential vorticity $Q$ and $\eta$ is assumed

$$\eta = \eta(x, y, Q, t), \quad (2.21)$$

which imposes the condition that also $Q$ varies monotonically with height. The tropopause is defined as the iso-PV surface, $\eta_{Q_0} = \eta(x, y, Q_0, t)$. Note that tropopause folds (or multitropopauses) are inconsistent with this hypothesis. They are presumed to be instantaneously
2.3. LOCAL EULERIAN METHODS

and irreversibly mixed, and are excluded à priori by considering only the highest tropopause surface (see Fig. 2.4). The trans-tropopause flux expressed with the generalised vertical coordinate is given by

\[ F(\rho) = \left[ \rho J_\eta \left( \frac{d\eta}{dt} - \frac{\partial \eta}{\partial t Q_0} - \mathbf{u} \cdot \nabla Q_0 \eta \right) \right]_{\eta Q_0} \quad (2.22) \]

where \( J_\eta \) is the Jacobian \( J_\eta = \| \partial z / \partial \eta \| \) of the vertical coordinate transformation. Basically, the vertical coordinate can be taken either as height, pressure, potential temperature or potential vorticity. With the vertical coordinate taken as \( Z \) (2.22) simplifies to the difference between vertical air motion and vertical tropopause motion. Wei (1987) suggested that the isentropic formulation (2.23) could better evidence the individual mechanisms leading to the exchange by claiming that one of the individual terms in (2.23) dominates for typical synoptic situations.

\[ F(\rho) = \left[ \rho J_\theta \left( \frac{d\theta}{dt} - \frac{\partial \theta}{\partial t Q_0} - \mathbf{u} \cdot \nabla Q_0 \theta \right) \right]_{\theta Q_0} \quad (2.23) \]

However, as discussed by Wirth (1995a), individual terms in (2.23) tend to act together and to cancel each other even in simple situations. For instance, the adiabatic advection of a cut-off low should lead to a zero exchange flux although terms 2 and 3 in (2.23) are non-zero. To avoid the cancellation problem and the interpretation ambiguity, Wirth (1995a) suggested to use the formulation with the PV vertical coordinate:

\[ F(\rho) = \left[ \rho J_Q \frac{dQ}{dt} \right]_{Q_0} \quad (2.24) \]

The use of this formulation requires computation of the material derivative of potential vorticity, which is a difficult task due to the derived, as opposed to observed, nature of PV.

Several attempts have been made to use the Wei-formulation for the quantification of STE, both on the global scale using assimilated data or on the synoptic scale using mesoscale model output data. Hoerling et al. (1993) did a calculation with the isentropic formulation
for the month January 1979 using 12 hourly ECMWF uninitialised analyses with two spatial resolutions, 3.75° and 1.875°, for both a thermal and a dynamical tropopause definition. The results differed from those provided by residual circulation estimates (cf. section 2.2): a net downward maximum flux around 30°-40°N and a distinct net upward flux maximum near 60°N, while tropical upward fluxes were found to be rather small in magnitude. In addition, cancellation between individual terms occurred over a significant range of latitudes. Lamarque and Hess (1994) applied the formulation with the PV vertical coordinate in a case study simulated with the mesoscale model MM4 in parallel to an other method based on the PV advection (see below). Model outputs with a spatial resolution of 80km, 27 levels were available every 3 hours. The Wei-formulation indicated small areas with very large values of mass exchange, in contrast to the advection method. They attributed this to the very high values of the Jacobian \( J_Q = |\partial z/\partial Q| \) in regions of an almost vertical 2PVU-tropopause. Siegmund et al. (1996) used the highest resolution (assimilated plus 3-hourly forecasts) data from ECMWF (3-hourly T213L31) to compute the STE flux with the Wei-formulation applied in pressure coordinate for the month of January 1994. They proposed an improved time derivation scheme which avoids the dipole structures resulting from the finite difference calculation of a spatial pattern which is advected with time. They obtained "realistic" large scale features, although their estimates were much larger than those diagnosed from the residual mean circulation. In their study, local and instantaneous fluxes are about 40 times as large as temporal and spatial averages, and therefore they suggested a high frequency of two way exchange. Again the study of Grewe and Dameris (1996) suggested strong two way exchange in the latitude belt 50°-70°N under the form of periods of large mass exchange from the troposphere into the stratosphere, disrupted by periods of mass exchange in the opposite direction. These two large opposite fluxes resulted into a much smaller net upward flux, subject to the uncertainties related to the cancellation. This study was based upon ECMWF T42 data on the comparatively long term period of 1979-1989, and used the isobaric Wei-formulation. Wirth and Egger (1999) tested the Wei-formulations in isobaric, isentropic and PV coordinate and compared them with a trajectory-based estimation in a case study simulated with the T106L31 ECMWF model (3-hourly outputs). They detected a similar tendency of the isobaric and isentropic formulations to create unrealistic small areas with large flux exchange resulting from the near cancellation of the scalar product \( \mathbf{u} \cdot \mathbf{n} \) where \( \mathbf{n} \) is the tropopause normal vector. Furthermore, in an adiabatic frictionless simulation, these two methods yielded exchange amounts of the same order of magnitude as found in the full-physics run. In contrast, the Wei-formulation using the PV coordinate and the trajectory-based method gave much better results, without significant flux exchange in the adiabatic simulation. Gettelman and Sobel (2000) made a climatology of STE using the Wei-formulation in isobaric coordinate using 6-hourly GEOS assimilated data at a resolution of 2°lat, 2.5°lon and 19levels. They did an error analysis and concluded that the large fluxes appear to be due to small amplitude events which might reasonably be explained as artifacts caused by errors or physical inconsistencies in the assimilated data. Consequently, they suggested that more reliable quantitative results can be expected when the method is applied to pure model outputs.

To summarise, the Wei-formulation is reliable only in its PV coordinate form. But the required material derivative of PV is difficult to extract from assimilated data, in opposition
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Table 2.2: STE estimated in case studies, in $10^9$ kg/s using different diagnostic methods. MM4: NCAR Mesoscale Model Version 8, ADV: advective method, see text.

to model output for which parameters involved in (2.4) are available. In its isentropic or isobaric form, the error of the Wei-formulation is of the same order of magnitude as the estimates themselves, although high resolution (temporal and spatial) and advanced time derivative schemes can significantly improve the results.

2.3.2 The PV Advection Formulation

An alternative Eulerian-based method to the Wei-formulation has been introduced by Lamarque and Hess (1994). It is based on the decomposition of the PV change due to advection and different physical processes and is set out as follows:

$$Q(t + \Delta t) = Q(t) + \left( \frac{\partial Q}{\partial t} \right)_{ADV} \Delta t + \left( \frac{\partial Q}{\partial t} \right)_{DIF} \Delta t + \left( \frac{\partial Q}{\partial t} \right)_{DIA} \Delta t$$  \hspace{1cm} (2.25)

Assuming that each term (ADV=advection, DIF=diffusion, DIA=diabatic) can be computed separately, the mass of exchange corresponding to diffusion can be estimated from the volume of air found between the two “tropopauses” where one “tropopause” is calculated with $\hat{Q}(t + \Delta t) = Q(t) + \left( \frac{\partial Q}{\partial t} \right)_{ADV} \Delta t$ and the other with $\hat{Q}(t + \Delta t) = Q(t) + \left( \frac{\partial Q}{\partial t} \right)_{ADV} \Delta t + \left( \frac{\partial Q}{\partial t} \right)_{DIF} \Delta t$. This method is not applicable to assimilated data because the estimation of each individual term is based on the knowledge of parameters which are only available from detailed model outputs. To render it applicable to assimilated data, it must be reduced to the form (2.26) (NA=non-adective) which does not provide a separation between diabatic and diffusive components any more. The exchanged mass can be estimated by comparing the purely advected PV field with the actual PV field at the next data time step.

$$Q(t + \Delta t) = Q(t) + \left( \frac{\partial Q}{\partial t} \right)_{ADV} \Delta t + \left( \frac{\partial Q}{\partial t} \right)_{NA} \Delta t$$  \hspace{1cm} (2.26)

This method has the advantage, when compared to Wei-formulation, that the full 3D structure of the tropopause can be resolved and it can be estimated from winds and PV fields which are available or can be derived from assimilated data. The reliability of this approach has however not yet been tested. It is to be expected for example that the 3D advection of the potential vorticity will be difficult to assess particularly where the tropopause possesses...
fine-scale structures. Furthermore, cancellation problems could appear when the purely advected PV field is spatially shifted compared to the PV field of the next time step. Such a shift is indeed not unlikely and attributable to either the 3D advection scheme or the data assimilation procedure.

### 2.3.3 A New and General Formulation of STE

A new Eulerian formulation is proposed here which overcomes problems encountered in the Wei-formulation at the vertical or folded structures of the tropopause, and should not be looked with strong cancellation effects. The estimates can be based on the available assimilated data set or on model output. Preliminary qualitative comparisons with the trajectory based method (see section 3) suggest the method to be promising. The analytical development is derived from the transport theorem, without restrictive assumptions, and is set out in Appendix A. The general result is:

\[ F(\rho) = -\int_S \rho \frac{1}{|\nabla Q|} \frac{DQ}{Dt} d\sigma \]  

(2.27)

The mass flux \( F(\rho) \) (positive downwards) across an iso-PV tropopause surface \( S \) depends on the gradient of potential vorticity, the material rate of PV and the tropopause surface area. The inverse dependency with the PV gradient confirms the common idea that exchange fluxes are reduced in regions of large PV gradients. The formulation can be seen as the generalisation of the Wei-formulation into a real 3D formulation and computation, and provides a conceptual interpretation of equation (3) of Wirth and Egger (1999) which they introduced as the basic principle for a new and much simpler development of the Wei-formulation.

The formulation which is regarded as adequate for assimilated data (see the diverse formulations in Appendix A) is:

\[ F(\rho) = -\int_S \frac{1}{|\nabla Q|} \left( \frac{\partial \rho Q}{\partial t} + \nabla \cdot (\rho Q \mathbf{u}) \right) d\sigma. \]  

(2.28)

The numerical algorithm consists in the local computation of the argument of the integration operator, followed by the 3D estimation of the local tropopause surface area \( d\sigma \). The PV gradient must be computed with its 3 components to avoid problems at locations with an almost vertical tropopause, and the time derivative should be computed using an improved scheme such as the one introduced by Siegmund et al. (1996). The local tropopause surface area can be estimated by a triangulation scheme, with particular care requested at locations where small-scale tropopause structures can invalidate the triangulation algorithm. Preliminary analyses indicate that the tropopause structure and its implication on the tropopause area accounts for a significant part of the exchange patterns, in contrast to what has been often assumed (for instance Wirth and Egger 1999).

### 2.3.4 Summary

The discussion in this section clearly shows the difficulties that can be expected when conducting a Eulerian diagnosis of local STE mass fluxes. An additional question concerning
these Eulerian methods is related to the accuracy of the tropopause location. The calculation of spatial derivatives for the potential vorticity tend to smooth the field, resulting in a concomitant smoothing of the estimated tropopause. This can induce significant errors in particular where the actual tropopause is expected to have fine structures. Quantitative estimates based on a local Eulerian method for selected case studies are summarised in Table 2.2.

To close the discussion on Eulerian methods, the particularly sensitive aspects are summarised hereafter. (1) The formulation must be computable from assimilated data. (2) The method should be tested with particular attention on assimilated data to assess the data inconsistency sensitivity. (3) Careful treatment of areas with almost vertical and multiple tropopauses because it is expected that most exchanges occur there. (4) Cancellation problem: orders of magnitudes of individual terms of the formulations should be analysed. (5) Time derivatives: dipoles structures resulting from time derivatives of timely sparse data could be avoided by a reliable time derivative scheme.

2.4 Lagrangian Methods

In a Lagrangian framework, observations or variables are looked at from a reference system which moves with an elementary volume of the atmospheric flow. This viewpoint can help the physical interpretation because changes along the flow often relate to basic physical processes. A simple and convincing example is the PV time variation: whilst $\frac{DQ}{Dt}$ directly furnishes information on non-conservative sources of PV (see (2.4)), the expression for $\frac{\partial Q}{\partial t}$ contains in addition the advective contribution. The observation of the Lagrangian rate of PV thus will directly furnish informations upon non-conservative sources of PV. Another reason for using a Lagrangian viewpoint relates to the chemical fingerprint of the flow. For instance, the water vapour content of an air parcel can modify locally the flow dynamics by exchanging thermal energy through radiation or latent heat release, and therefore knowledge of the history of the parcel given along its trajectory can provide an indication of such heating sources (Wernli and Davies 1997). On the other hand, the flow can induce a “remote” chemical forcing through exchange between two zones where chemical conditions are different. Again, trajectory analysis can provide a helpful tool to diagnose such events. In particular, trajectories have been widely used in past studies to identify the origin of high ozone concentrations measured in the troposphere and to prove that stratosphere-troposphere exchange has taken place (Danielsen 1980, Ancellet et al. 1991, Vaughan et al. 1994, Rood et al. 1997, O'Connor et al. 1999, Eisele et al. 1999). At larger scales, the Lagrangian viewpoint is often realized by studying the motion of tracers using transport codes. In essence these transport codes advect elementary volumes with given 3D winds, usually using a comparatively complicated numerical scheme. They can be coupled with a chemistry model (chemistry transport models CTM) and can provide global chemistry information based either on assimilated winds (Chen 1995, Velthoven and Kelder 1996) or on the output winds of a general circulation model (GCM) (Stone et al. 1999). Velthoven and Kelder (1996) estimated quantitatively the STE flux across the 100hPa surface using a transport code coupled with chemistry modules based on ECMWF analyses with several
relatively coarse resolutions $(8^\circ \times 10^\circ, 4^\circ \times 5^\circ, 2.5^\circ \times 2.5^\circ)$. They show a large dependency of the diagnosed mass fluxes to the resolution, and furthermore a sensitivity to changes in the model used for the data assimilation (see Table 2.1). Transport codes permit the study of the temporal evolution of the distribution of an artificial tracer, but they are usually not able to provide useful trajectories. Thus, trajectory computation codes are sometimes required on the global scale to complement transport models (for instance Pierrehumbert and Yang 1993, Sparling et al. 1997).

On the other hand, there have been only few attempts to look at exchanges using systematically computed trajectories. In perhaps the first such study Wernli and Davies (1997) identified exchange events in the context of a real case study and provided a quantitative STE estimate which was comparable to those of Lamarque and Hess (1994) (see Table 2.2). The procedure was based on a discretisation of the flow and the computation of the correspondent trajectories. Another quantitative Lagrangian estimate of STE has been given by Wirth and Egger (1999) in a comparative study between Eulerian methods following Wei (1987) and a trajectory based method (as discussed in section 2.3). They concluded that together with the Wei-formulation in PV coordinate, the trajectory method was the most accurate. They used a different scheme than Wernli and Davies (1997) which corresponds to a Lagrangian realization of the advective method of Lamarque and Hess (1994) described in the last section: starting trajectories on the tropopause, the volume enclosed between the advected tropopause and the actual new tropopause was regarded as the exchanged mass. However, with output from their adiabatic frictionless run they obtained a downward flux half as large as for the full-physics run, and the upward flux was as large. This shows that although this particular Lagrangian method was as reliable as the best Eulerian method, it is again subject to significant uncertainties, probably in connection with the problems discussed in the last section.

2.5 Summary

In summary the discussion of this Chapter has shown that the existent methods possess significant uncertainties. Strong cancellation problems occur with Eulerian methods using the Wei-formulation in isobaric or isentropic coordinate. Comparatively more confidence can be placed upon the Wei-formulation cast in a PV coordinate framework and from the trajectory based methods. However, the lack of “true” (observational) estimates leaves the inter-comparison as the only feasible way of testing the quality of a method. Moreover, on the global scale, geographically resolved quantitative STE estimates have so far only been calculated with methods that have been shown less reliable in the synoptic-scale studies. Probably more realistic are global estimates provided by methods based upon the residual mean circulation. To conclude, it can be stated that (1) present STE estimates must be interpreted with caution and knowledge, (2) there is evidence that trajectory-based methods can yield fair STE estimates, (3) any proposed method should be analysed in detail and with care to avoid increasing the profusion of STE estimates which are as uncertain as others already existent.
Chapter 3

Development of the Lagrangian Method in a Case Study

3.1 Preamble

The earlier discussion of various diagnostic methods for the quantification of STE indicates that the Lagrangian framework has significant potential.

This Chapter is geared to developing and testing a Lagrangian method for the quantification of STE, and is an attempt to provide a reliable method for the diagnostic of STE. The strategy is first to characterise the numerical representation of the cross-tropopause exchange event in a Lagrangian framework, and to assess the exchange event with a realistic model (hereafter, referred to as the "exchange event model"). Then, application of the exchange event model to a synoptic situation is done with particular attention being given to optimising the exchange event model's parameters and to assessing its possible limitations.

The meteorological context is a synoptic development which has been chosen for its similitude to the typical synoptic development that is recognised to contribute for the largest part of the overall stratosphere-to-troposphere exchange in the mid-latitudes (Chapter 1). The calculations are based upon high spatio-temporal resolution data (0.5° x 0.5°, 1 h) provided by a mesoscale model.

The estimates of STE yield by applying the method to the same synoptic development are discussed in the next Chapter with two underlying goals: (i) evaluate the method; and (ii) analyse the physics and dynamics of exchange. Then, the method is further applied in the subsequent Chapters to derive and analyse a one-year hemispheric Lagrangian climatology of STE.

3.2 Basic Numerical Algorithms and their Errors

From our Lagrangian viewpoint, an exchange occurs when an air parcel crosses the tropopause, and the mass flux across the tropopause is quantified by the number of parcels which cross the tropopause. As detailed below, the air parcel is numerically traced by trajectory computation and the tropopause determined by computing the 2PVU iso-surface. It is clear that
both the computation of trajectories and the tropopause are contaminated by numerical errors. This can lead to systematic biases in the quantification of STE if each event where a trajectory crosses the tropopause is considered as exchange event (Sobel et al. 1997). It is therefore important to take account of numerically induced errors in developing a method for estimating STE.

3.2.1 The Trajectory Computation Algorithm

The trajectories are computed with the three-dimensional Lagrangian computation code Lagranto (Wernli and Davies 1997) based on Petterssen’s kinematic method (Petterssen 1956). Each trajectory step \((t \rightarrow t + \Delta t)\) is evaluated with the iterative scheme:

\[
\begin{align*}
    r_1 &= r_0 + \Delta t u(r_0, t) \\
    r_i &= r_0 + \frac{\Delta t}{2} (u(r_0, t) + u(r_{i-1}, t + \Delta t)), \quad \text{for } i \geq 2
\end{align*}
\]

where the \(r_i\) are the iterative evaluations of the trajectory position at \(t + \Delta t\) which are assumed to converge to the exact position \(r(t + \Delta t)\)

\[
\lim_{i \to \infty} r_i = r(t + \Delta t)
\]

and \(u\) is the three-dimensional wind field.

The trajectory time step \(\Delta t\) is a crucial parameter for the accuracy of the trajectory. Seibert (1993) studied the convergence and accuracy of trajectories computed with Petterssen’s method for several idealised wind configurations. It was shown that convergence (3.3) occurs only if the time step \(\Delta t\) is smaller than a limit \((\Delta t < \Delta t_c)\) which depends on the wind structure (vorticity, shear, acceleration). Furthermore the accuracy of trajectory computation is sensitive to the time step \(\Delta t\) such that an adjusted upper limit corresponding to about 15\% of \(\Delta t_c\) is necessary. The final choice of \(\Delta t\) depends on the nature of the wind distribution and it has been suggested thus a time step \(\Delta t \leq 30\) min. The trajectory time step \(\Delta t\) in Lagranto is set to 1/12 of the data temporal resolution, which corresponds to 5 min in the case study presented in this Chapter (model data every 1 h). This choice is therefore expected to provide a reasonable accuracy even in extreme wind configurations.

On the other hand, the code Lagranto does not test explicitly the convergence achievement (3.3) but it is assumed after three iteration steps. Seibert (1993) showed that for time steps \(\Delta t = 6\) h the part of converging trajectories after 3 (6) iterations is about 75\% (99.5\%) in the troposphere. For time steps \(\Delta t = 5\) min, it is reasonable to expect a much quicker converging rate and therefore a ratio of converging trajectories close to 100\%.

Doty and Perkey (1993) studied the accuracy of trajectory computation using the Petterssen’s scheme with trajectory time steps of 5 min calculated within two iterations. They showed that the temporal resolution of data has no significant impact on the trajectory accuracy if it remains higher than 3 h. The discussion at lower temporal resolutions is provided in the Appendix C when the sensitivity to the data resolution will be addressed.
3.2.2 Determination of the Tropopause

The tropopause is numerically determined by the computation of the potential vorticity three-dimensional field and the identification of the 2PVU iso-surface\(^1\). The calculation of potential vorticity via (2.5) is a non-linear operation which involves the product of derivatives of the wind and temperature fields. Derivatives are numerically evaluated at each grid point from the finite difference of values at the two nearest grid points. It should be noted that this derivation scheme can lead to local inconsistencies of the derivated field because the calculations of two neighbour values do not imply any common information. Such local inconsistencies in the PV field are likely to occur in regions where significant changes in the winds occur between two neighbour grid points, and are to be expected in spatially complex regions such as upper-level frontal systems and tropopause folds.

On the other hand, the effect of the finite data resolution on the final 2PVU iso-surface is a general spatial smoothening with a possible additional vertical shift. For instance Wernli (1995) (their Fig. 2.4 and 2.5) evidenced the sensitivity of the PV field to the data vertical resolution within a synoptic situation by comparing PV fields computed from data with 31 and 15 levels. Differences in the tropopause vertical location were found to reach 50 hPa in some regions.

3.2.3 Calculation of PV along Trajectories

The identification of cross-tropopause events requires knowledge of the PV evolution along the trajectory. For the trajectory time steps where the wind and temperature fields are available (around the trajectory position), a bilinear spatial interpolation is performed. For trajectory time steps where these fields are not available, a temporal interpolation must be combined with the spatial one. This combination must be undertaken carefully due to the non-linear nature of the PV calculation.

Figure 3.1 shows values of potential vorticity along a trajectory as calculated following two different temporal interpolation schemes. The trajectory during its first 24 h lies within a domain where data is available every 1 h, and then enters a domain where data is available only every 6 h. The local linear temporal interpolation of the three-dimensional PV field every 1 h induces unrealistic\(^2\) large oscillations in the PV values. This scheme can therefore not be applied. To ensure that such artificial oscillations are not introduced, the pseudo-materiologically conserved nature of potential vorticity has been used and the following scheme has been chosen: PV is calculated from wind and temperature data only at trajectory time steps where these fields are available, and a material linear temporal interpolation is performed for the intermediate trajectory points. This has the effect of removing the unrealistic oscillations as shown by the dotted line in Fig. 3.1.

\(^1\)A preliminary discussion of the choice of the tropopause as the 2PVU iso-surface is provided in Chapter 1.

\(^2\)These oscillations have a frequency corresponding exactly to the temporal resolution of the data.
3.2.4 Implications Related to Cross-Tropopause Events

The foregoing highlighted errors in the computation of either trajectories or tropopause have implications for the “activity” of cross-tropopause movement.

On the one hand, trajectories with convergence problems near the tropopause can easily induce spurious cross-tropopause events. However, based on the preceding discussion, it is reasonable that in the present high resolution case study, such events are rare in comparison to realistic cross-tropopause events.

On the other hand, the errors in the numerical determination of the tropopause can lead to systematic errors. The local inconsistencies of the PV field induce localised abnormal PV values along trajectories. The non-biased smoothing of the tropopause makes trajectories oscillate about the tropopause. These two errors spuriously increase the cross-tropopause activity and would lead to a systematic over-estimation of the STE if not handled. To allow for this deficiency a discriminator, the *residence time criterion* is introduced in the next section. A possible bias in the vertical location of the tropopause would lead to evaluating mass fluxes at the incorrect location and correction of such an error is, however, difficult and it is assumed that the high spatial resolution used in this case study ameliorates this effect.
3.3 The Lagrangian Model of the Exchange Event

3.3.1 Discriminating the Numerically-Induced Trans-Tropopause Movement: The Residence Time Criterion

The main methods for the purpose of discriminating the purely numerically-induced events from the significant one are based either on a finite width tropopause transition layer, or on the material residence time before/after the exchange.

From the first viewpoint, the tropopause is seen as a transition layer between the stratosphere and the troposphere, and only trajectories going from one side of this layer to the other side are considered “exchanged”. The layer can be defined for example by the points closer than a given distance $\Delta D$ (in m) from the 2PVU surface, or by the two iso-PV surfaces $2 \pm \Delta PV$, respectively. In the first case, the layer is composed of zones with small mixing activity as well as zones with very high mixing activity (for example jet stream regions). The choice of an overall optimised distance $\Delta D$ is therefore difficult and the amount of lost “real” exchanges can be very sensitive to this choice. In the other case, the two iso-surfaces $2 \pm \Delta PV$ are affected by the same numerical problems as the 2PVU iso-surface, and therefore the determination transition layer also suffers from numerical inconsistencies and smoothing. The use of such a discriminator is likely to introduce additional problems in the regions where the tropopause has small-scale structures like for instance upper-level fronts.

From the second viewpoint, the exchange trajectories are selected with a criterion based upon the time they spent within the troposphere and within the stratosphere, before / after the exchange. These so-called residence times are required to be longer than a critical residence time $\tau_{crit}$ in order to consider the parcels as having had a significant interaction within the respective atmospheric layer. Trajectories that experience tropopause crossings shorter to each other than the critical residence time $\tau_{crit}$ are not selected. This residence time method minimises the use of the PV field to the determination of the tropopause. Moreover, chemical and radiative forcing in the troposphere (respectively the stratosphere) linked to the exchange is directly related to the exchange parcel’s residence time within the troposphere $\tau_t$ (respectively the stratosphere $\tau_s$). Hence the neglected, but realistic, exchanges with residence times smaller then $\tau_{crit}$ can be considered as having a relatively small impact if $\tau_{crit}$ is small enough.

In this study the residence time selection criterion has been adopted because it appears to optimise the degree to which the numerically-induced crossings can be excluded, and at the same time the relative importance of the eliminated “exchanges” is small and under control.

3.3.2 The Transition Time Period

An exchange model can be portrayed schematically in a manner consistent with the idea of a critical residence time (Fig. 3.2): an exchange between the troposphere and the stratosphere will be considered only if the trajectory experiences a journey of at least a period of $\tau_{crit}$ in both the troposphere and the stratosphere, consecutively.
The remaining question concerns the transition time period $T^{\text{trans}}$ between the two residence time periods. On the one hand, consistent with the preceding discussion, trajectories must be allowed to experience multiple crossings while going through the tropopause. But on the other hand, if the transition period is too long, a trajectory could spend a long time near the tropopause and change its chemical properties there. To give emphasis on the events which induce interaction between the stratosphere and the troposphere, the maximal transition period $T^{\text{trans}}_{\text{crit}}$ must be taken smaller than the critical residence time $\tau_{\text{crit}}$.

### 3.3.3 The Exchange Location

A consequence of allowing a non-zero transition period is that location of the exchange becomes a question of convention. Methods distributing the exchange point into several partial exchange points inside the transition period have not been considered to avoid interpretation difficulties.

Our choice has been to select the last tropopause crossing point of the transition period, as the "incoming point" where the exchanged air mass enters the new atmospheric layer. In Appendix B.4 geographical exchange distributions computed with this convention are compared with maps where the converse "outgoing point" has been used. The sensitivity is found to be negligible.

### 3.4 The Numerical Set Up

Hereafter the exchange event model (Fig. 3.2) is applied to a case study with the aim of providing a quantification of STE of a relatively high resolution.

#### 3.4.1 Data Set

To produce a high-resolution data set data from the European Centre for Medium Range Weather Forecast (ECMWF) are combined with output from a mesoscale hindcast simula-

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3The effect of the transition time on the exchange estimates is discussed in the Appendix B.2.
This simulation has been performed for a duration of 162 h starting on Sept 1 00UTC and ending on Sept 7 18UTC using the Europa Model (EM) from the German Weather Service (Majewski (1991)) driven by six-hourly ECMWF analysis fields (T213L31). The simulation domain is represented in Fig. 3.4 by the bold line, and the resolution is 0.5° in the horizontal and 40 levels in the vertical (levels every 20 hPa in the tropopause region between 150 and 510 hPa, and every 30 hPa elsewhere). The simulation results compare reasonably well with the ECMWF verifying analysis, and reproduce correctly the different phases of the development both in amplitude and location. For the purpose of quantifying STE, a diagnosis domain has been chosen over western Europe to encompass the whole baroclinic development. This domain is represented by the gray shaded sector in Fig. 3.4. However, data outside the diagnosis domain is required to enable trajectories time-scales to exceed the residence time criterion. Therefore, ECMWF analysis and EM simulation data sets have been merged to form a homogeneous hemispheric data set, whose grid resolution corresponds to the one of the EM simulation (0.5°, 40 levels, 1 h). All required spatial (horizontal and vertical) and temporal interpolation is proceeded using linear schemes.

Finally, for special issues involving other data resolutions, such as the study of the sensitivity of estimates to the data resolution (Appendix C), the required resolution (Table 3.1) is produced from this homogeneous data set spatially via linear interpolation and temporally
3.4.2 The Three Step Computation of Exchange Trajectories

The exchange trajectory set is obtained by a three step computation scheme. First, the time-dependent flow is discretized into a set of trajectories with limited length, filling the whole three-dimensional domain of interest. Secondly, they are prolonged to allow the application of the residence time criterion, and in a third step, the residence time and transition period criteria are applied.

**Step 1: Lagrangian air flow representation**

The time-evolution is split into consecutive time-periods of length $T$, and the air flow is discretized for each period by a set of trajectories of length $T$ starting on a three-dimensional regular grid, hereafter the *starting grid*, such that each trajectory represents the evolution of an elementary air volume during $T$.

To produce a full representation of the air flow around the tropopause within the diagnosis domain for the entire period $T$, the starting grid must have a sufficient extent to account for the overall horizontal and vertical advection during $T$. The determination of the starting grid box has been done by starting backward trajectories in the tropopause region for a period $T$ and by choosing the domain which encompass at least 95% of the ending points of these trajectories. The quantitative results for several choices of $T$ are given in Appendix B.1, and the box corresponding to the configuration chosen here is represented in Fig. 3.4 and extends vertically between 600 and 50 hPa. It leads to less than 1% of unrepresented parcels in the tropopause region.

The elementary volumes, and therefore the resolution of the *starting grid*, must be chosen such that counting centres of elementary volumes crossing the tropopause gives a good approximation of the volume crossing the tropopause, even on small scale. The optimisation of the horizontal and vertical size of the elementary volumes is discussed in detail in Appendix B.1. It turns out that a value of $T = 12$ h and a starting grid with a horizontal resolution of 0.5° and a vertical spacing of 5 hPa, suffice to give a reasonable representation of the temporal structure of exchange patterns and simultaneously permits the parcels to be advected far enough from their starting points to expect a reasonable filling of the volume, both in horizontal and vertical directions. An increase of the horizontal resolution from 0.5 to 0.25°

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4E.g. only required times are used
only weakly improves the quality of the results but would multiply the computation time by a factor four.

The trajectories are launched from the grid points of the starting grid for each consecutive period of length $T$. A preliminary trajectory selection is operated at this point by keeping only those which intersect the tropopause. Those are potential candidates for our exchange model and are therefore called *preliminary exchanges*.

**Step 2: Trajectory prolongation**

Although exchange events occur within the time period $T$ discussed above, our exchange selection model requires further information on the trajectories’ past and future. They are extended backwards for a period $T^b$ and forwards for $T^f$, yielding a total integration time of $T + T^b + T^f$. The choice of $T^f$ and $T^b$ is directly related to the choice of the critical residence time $\tau_{\text{crit}}$ and transition period $T^\text{trans}_{\text{crit}}$ (that is, $T^b > \tau_{\text{crit}} + T^\text{trans}_{\text{crit}}$, and $T^f > \tau_{\text{crit}}$).

Here, trajectory prolongation has been computed for 120 hours to allow a residence time sensitivity study in the next Chapter. Such a trajectory length is generally regarded as yielding acceptable accuracy.

A delicate issue related to the trajectory prolongation is the resolution change of the wind fields at the borders of the limited-area model and related model boundary effects. Exchange events are investigated only inside the diagnosis domain, and trajectory portions outside the domain are only required to apply the residence time criterion. Thus, the accuracy requirements outside the diagnosis domain are less than inside, and restricted to cross-tropopause activity. Detailed discussion (Appendix B.3) shows that the only significant effect is the reinforcement of non-conservative forces at the lateral boundary of the simulation due to the relaxation boundary condition in the model. This effect is however estimated to influence less than 5% of the exchange events.

**Step 3: Selection of significant exchange events**

Finally, the significant exchange events are selected in accordance with the exchange event model (cf Fig. 3.2).

The selection depends on two parameters which are the critical residence time $\tau_{\text{crit}}$ and the critical transition period $T^\text{trans}_{\text{crit}}$. A value of $\tau_{\text{crit}} = 12\,\text{h}$ is used in this study, conforming to the analysis given in Appendix B.2 where it is shown that the cross-tropopause activity is actually highly dependent of data resolution only for residence times smaller than $12\,\text{h}$ (see Fig. B.12). This behaviour provides a further *a posteriori* justification for the use of the residence time criterion to reduce numerically-induced errors. The critical transition period is taken as $6\,\text{h}$ (Appendix B.2).

### 3.4.3 Exchange Mass Quantification

Assuming the mass to be conserved along the trajectories, the mass represented by each trajectory is determined by the starting grid resolution and can be represented at the exchange location. Due to the special choice of the starting grid, the mass of the elementary volume $\Delta M$ is constant. Using the hydrostatic relation $\Delta M$ can be written as

$$\Delta M = -\frac{1}{g}(\Delta x)^2 \Delta p$$  

(3.4)
where, \( g \) = the constant of gravity,
\( \Delta x \) = horizontal resolution of the starting grid,
\( \Delta p \) = vertical resolution of the starting grid.

3.5 Summary

The analysis of the Lagrangian approach of quantifying STE has led to define a model for a realistic exchange event based on the residence time criterion. The residence time criterion has been introduced to eliminate events induced by numerical errors.

The exchange model’s parameters have been established. The smallest critical residence time is \( \tau_{crit} = 12 \text{ h} \), and a critical transition time \( T_{crit} \approx \frac{1}{2}\tau_{crit} \) has been suggested (Appendix B.2).

The starting grid determining the discretisation of the atmospheric flow has been analysed and optimised for the purpose of a case study, and for a range of data resolutions (Appendix B.1).
Chapter 4

A Detailed Case Study Analysis

In this Chapter, estimates of stratosphere-troposphere exchange performed using the methodology discussed in the previous Chapter are presented and analysed. Two goals are pursued: the evaluation of the method and the physical analysis of cross-tropopause exchange.

The quantification of STE in a prototypical baroclinic development permits to gain insight into the typical mechanisms responsible for most stratosphere-to-troposphere exchange in the midlatitudes. Furthermore, the high resolution and the Lagrangian nature of the method enable a detailed analysis of these mechanisms. In turn, the detailed analysis of STE estimates allows the qualitative verification of the results and provides an evaluation of the method.

The development occurred over western Europe between Sept 1 and 5 1997. It is the late stage of a typical baroclinic wave development corresponding to the anticyclonic-shear case described by Davies et al. (1991), Thorncroft et al. (1993) and Wernli et al. (1999). The time window has been chosen to include the baroclinic wave breaking and the decay of the resulting cut-off, because these processes are expected to be associated with large exchange.

The temporal evolution of the geographical distribution of exchange is presented first and discussed with regard to the temporal evolution of the tropopause. The exchange is further analysed in a three-dimensional perspective, and physical interpretations of the diagnosed exchange patterns are set out for two identified major exchange episodes. Finally, exchange estimates are discussed with regard to the considered iso-PV level, and to the critical residence time.

4.1 Tropopause Evolution and Exchange

Figures 4.1 and 4.2 (left panels) represent the evolution of the potential temperature on the 2PVU iso-surface. This two-dimensional representation of the tropopause gives some indication of the vertical structure of the tropopause, but does not provide information on tropopause folds (only the highest level of the tropopause is actually displayed). The information provided by this representation is similar to that given by isentropic charts of potential vorticity, and a similar dynamical interpretation can be adopted.
Figure 4.1: Time evolution of the tropopause and exchanged mass. Left: time evolution of θ [K] on the 2PVU surface, at (a) Sept 1 06UTC, (b) Sept 1 18UTC, (c) Sept 2 06UTC, (d) Sept 2 18UTC. Right: net exchange mass [$10^{12}$ kg] per grid-square (1°x1°) integrated over time periods of 12h (A) Sept 1 00-12UTC, (B) Sept 1 12-24UTC, (C) Sept 2 00-12UTC, (D) Sept 2 12-24UTC. Downward mass fluxes are positive. Bold lines represent the 325 and 330 K potential temperature contours on the 2PVU surface.
4.1. TROPOPAUSE EVOLUTION AND EXCHANGE

Figure 4.2: As Fig. 4.1. Left: (e) Sept 3 06UTC, (f) Sept 3 18UTC, (g) Sept 4 06UTC, (h) Sept 4 18UTC. Right: (E) Sept 3 00-12UTC, (F) Sept 3 12-24UTC, (G) Sept 4 00-12UTC, (H) Sept 4 12-24UTC.
ANALYSIS

Figure 4.3: Evolution of the hourly exchange mass for the period 12UTC to 24UTC of the 1st of September within the zone of large downward exchange associated with the streamer's break-up process (see Fig. 4.1 B). Exchange mass is in units of $10^{12}$ kg h$^{-1}$ for the domain 5W-0E, 39N-50N.

Figures 4.1 and 4.2 (right panels) depict the estimated exchange masses between Sept 1 00UTC and Sept 4 24UTC. Here, the 12-hourly accumulated net exchange masses have been represented instead of the separated values for upward and downward exchange because this representation gives a more direct picture since the two exchange directions occur in geographically separated regions on the considered time scale of 12h.

The general structure of exchange patterns reveals well identifiable zones of strong exchange activity, and a relatively weak "background". The active zones are confined in space and time and occur almost exclusively in the region of the stratospheric intrusion at some specific stages of its development.

The elongated north-south oriented streamer arrives over western Europe and begins to break on Sept 1 around 12UTC. It takes about 3 days to break up all isentropic contours for values below 340 K on the 2PVU tropopause. However, most of the tropopause irreversible deformation associated with the break-up occurs between Sept 1 06UTC and 2 06 UTC (Fig. 4.1, (a) - (c)). This leads naturally to the strong stratosphere-to-troposphere exchange during the period Sept 1 12UTC - 24UTC (see Fig. 4.1, (B)). The exchange pattern is composed of two distinct patches of strong downward exchange. The northern one corresponds to the streamer's break-up region and the southern one is located at the tip of the streamer.

The detailed hourly development of this major exchange episode between 12 and 24UTC of Sept 1 is represented in Fig. 4.3. It reveals that most of the exchange attributed to the streamer's break-up process occurs within a time period of 6 hours. The temporal and spatial confinement of this explosive event emphasises the mesoscale nature of STE. The complex structure of the tropopause during this period and the related exchange is further discussed below from a three-dimensional perspective. The later stages of the break-up are much smoother and imply weaker downward exchanges.

The break-up of the streamer closes the lowest isentropic contours and progressively forms a cut-off system. The cut-off can be regarded as a dynamical entity from Sept 3
4.1. TROPOPAUSE EVOLUTION AND EXCHANGE

Figure 4.4: Evolution of the hourly exchanged mass for the period Sept 3 00UTC to 4 24UTC in the zone of large downward exchange associated with the cut-off (in $10^{12}$ kg h$^{-1}$ for the moving domain [0E-10E x 40N-50N] to [5E-15E x 37N,47N]). Labels (E) to (H) refer to the panels of Fig. 4.2.

06UTC onwards (see Fig. 4.2 (e)). It enters immediately a decay phase and has almost entirely disappeared by Sept 5 00UTC. At the bottom of the cut-off weak troposphere-to-stratosphere exchange occurs only at the beginning of this period (Fig. 4.2 (E)). Afterwards the decay is associated purely with stratosphere-to-troposphere exchange. The downward exchange zone exhibits a high coherency. It starts on the border of the cut-off and tends to focus progressively toward the centre. Figure 4.4 shows the detailed temporal evolution of the net downward exchange during the decay of the cut-off. The cut-off experiences a strong loss of stratospheric air on Sept 3 between 08UTC and 14UTC, and then the exchange rate reduces but with large oscillations until Sept 4 12UTC when it drops to very small values. Here again, the exchange geometry and the related cut-off decay involve fine-scale structures which are fundamental for the physical understanding of cross-tropopause exchange and further analysis is provided in the next section from a three-dimensional perspective.

Beside these major exchange episodes some other weaker events also occur. A first example appears east of the streamer (Fig. 4.1 (c)) where a patch of locally raised tropopause height decays irreversibly during the next 24 hours. This decay is associated with the troposphere-to-stratosphere exchange patch east of the streamer in panels (C) and (D). Another event is the northward tongue of upward exchanges to the west of the “stretching” streamer (see Fig. 4.2 (F)). This is associated with the decay of the thin “negative streamer” which is visible in this region on panel (f).

\[^{1}\text{A negative streamer denotes here the analogue to the streamer but in association with a negative PV anomaly.}\]
4.2 Three-Dimensional Perspective

Complex three-dimensional structures of the tropopause are often associated with localised exchange events. To examine the relationship between tropopause deformation and exchange, it is illuminating to consider the structure of the tropopause and the associated “exchange geometry” from a three-dimensional perspective. Time sequences of selected three-dimensional views are provided in Figs. 4.5 and 4.6 for the streamer’s break-up phase and the cut-off decay, respectively. The tropopause is represented by the blue surface and sectors of exchange trajectories are drawn as red (green) traces for stratosphere-to-troposphere (troposphere-to-stratosphere) exchange.

Five stages of the explosive break-up are presented in Fig. 4.5: (a) well before (10UTC), (b) at the beginning (14UTC), (c) at the maximum (15UTC), (d) towards the end (16UTC), and (e) well after (23UTC) the event. The temporal evolution of the tropopause compares very well with the exchange evolution already shown in Fig. 4.3, where in particular the exchange maximum was found to occur at 15UTC. Moreover, stratosphere-to-troposphere exchanges are located at the places where the deformation of the tropopause is strong. At 10UTC (Fig. 4.5 (a)), the tropopause intrusion is shaped as a tilted “funnel” bounded by an almost vertical wall on the southern side which rolls up cyclonically. Then, between 14UTC and 16UTC, the “isentropic bottom” is rapidly eroded. An arched structure is progressively formed, leading to a subsequent intrusion at the northern boundary and an isolated vertical tube at the southern boundary (panel (d)). And finally (Fig. 4.5 (e)), the lower part of the tube is entirely mixed and the tropopause resembles its initial state but with a much shallower intrusion.

The decay of the cut-off on Sept 3 is also represented in three dimensions in Fig. 4.6. The cut-off\(^2\) is progressively split and an isolated blob is formed. This splitting occurs in a thin isentropic layer near 328K and is clearly associated with stratosphere-to-troposphere exchange. The subsequent progressive decay of the isolated blob is also associated with stratosphere-to-troposphere at its bottom. Note that the disappearance of the isolated blob coincides with the eventual reduction of the exchange flux by Sept 4 12UTC found in Fig. 4.4.

4.3 Analysis of Physical Processes

The exchange has been shown to possess fine structures consistent with tropopause deformation. In this section, an attempt is made to qualitatively diagnose the accompanying physical processes.

The material change of potential vorticity \( Q \), which leads to exchange, is driven by the diabatic heating \( \dot{\theta} \) and friction/turbulence \( F \) fields:

\[
\frac{DQ}{Dt} = \frac{1}{\rho} \{ \omega \cdot \nabla \dot{\theta} + \nabla \wedge F \cdot \nabla \theta \}.
\]  

\(^2\)Here the denomination “cut-off” always denotes closed isentropic PV contours, as opposed to a “blob” which is thought as a three-dimensional closed surface.
4.3. ANALYSIS OF PHYSICAL PROCESSES

Figure 4.5: Three-dimensional view of some selected instants of the break-up phase on Sept 1. The blue surface is the 2PVU tropopause (in the domain 15W-15E, 35N-58N, and vertically from 4 to 16 km). Red (green) traces are portions of stratosphere-to-troposphere (troposphere-to-stratosphere) exchange trajectories. The traces are composed of three consecutive trajectory sectors of an hour, the central one corresponding to the exchange location. (a) at 10UTC, view from the south-west, (b) at 14UTC from south-east, (c) at 15UTC from south-east, (d) at 16UTC from south-west, (e) at 23UTC from south-west.
Figure 4.6: Same as Fig. 4.5 but for the cut-off decay phase on Sept 3 and 4. View from the south. (a) 3 at 08UTC, (b) 3 at 12UTC, (c) 3 at 20UTC, (d) 3 at 23UTC, and (e) 4 at 10UTC
The availability of the two non-conservative sources directly from the numerical model would permit to identify the driving forces at each location. However these fields are not available directly from the model, and their computation/estimation from the basic fields \((T, u, q)\) involve a sequence of numerical derivations and interpolations which act as a scale filter. When computed this way, the diabatic heating rate and its gradient or a turbulence indicator such as the Richardson number certainly miss very small-scale structures, and therefore can not be used as comprehensive indicator of exchange where such small scales are involved. For this reason, the analysis provided hereafter is based upon the available fields \((T, u, q)\) and derived fields involving little numerical scale filtering.

The “total relative humidity”, the ratio between vapour pressure of the total water content and saturation vapour pressure evaluated with respect to water / supercooled water, is used to trace moisture. The reason to use this quantity instead of the specific humidity (the “true” tracer) is related to: (i) the very low values of specific humidity in the tropopause region compared to those in the lower troposphere; and to (ii) its use as indicator of the cloud formation conditions. Furthermore, the cloud water content is used to indicate the presence of clouds and related effects, and the partial cloud cover is used to indicate the presence of enhanced radiative effects.

### 4.3.1 The Streamer Break-up Episode

The break-up of the streamer (see Fig. 4.5) takes place principally between 14 and 16UTC of Sept 1, and coincides with an intense cumulus cell east of the streamer and north of the Pyrenees. This cloud cell has an embryonic shape at low levels at 08UTC and strengthens and grows vertically as the streamer is advected eastwards. It attains a maximum size between 14 and 17UTC and decays very rapidly at 18UTC. The left panel of Fig. 4.7 represents the cloud cell at 14UTC. The cumulus formation is triggered by the northward and upward advection of humid air driven by the cyclonic circulation induced by the streamer itself, and is probably reinforced by passage over the Pyrenees. The cumulus is associated with strong condensational heating in the interior and with radiative cooling at the top. An indirect indicator of the presence of strong diabatic heating within the cloud cell is the zone of very low potential vorticity values \((Q \sim -0.5\text{PVU})\) which is found in the outflow region of the cloud cell, very close to the tropopause level. This is due to the strong ascent of parcels coming from low tropospheric levels (typically with low PV values) within the cloud.

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3 A new version of the EM model, the High-Resolution Model (HRM) (see Majewski (1991)) has been modified successfully to provide all non-conservative sources intervening in the momentum equation and would therefore be more appropriate for quantitative analyses of physical processes responsible for exchange. The use of this model will be a possible further development of this thesis.

4 \(q\) is the total water content.

5 In the model, the prognostic variable related to water is the total water content (without distinction between vapour and condensed water). Clouds are formed as soon as the total water content surpasses the saturation vapour pressure over water (at temperatures below 0°C the saturation vapour pressure is evaluated with respect to supercooled water and not with respect to ice).

6 A partial cloud cover is performed in the model if the vapour pressure reaches values greater than a threshold which is typically 80% - 90% (at tropopause levels, depending on the pressure) of the saturation vapour pressure evaluated with respect to either water or ice. Partial cloud cover is not subject to the cloud microphysic of the model but has enhanced radiative effects in comparison to pure water vapour.
region which implies first an increase of $Q$ as the diabatic heating vertically increases near the cloud bottom and then an even stronger decrease of $Q$ at levels above the maximum diabatic heating rate (Wernli and Davies 1997, Pomroy and Thorpe 2000). A zone of very low PV values close to the tropopause acts to strengthen the PV gradient and in that way is likely to reinforce the jet stream locally. This effect can be seen very clearly on a sequence of charts representing the horizontal velocity at 300 hPa (not shown): a strong local reintensification of the jet stream, namely a jet streak, reaching a maximum of 50 m s$^{-1}$, takes place right downstream of the cloud cell along the streamer.

The south-western part of the cloud cell is located below the streamer near the region of explosive erosion, as it is apparent at the 350 hPa level in the right panel of Fig. 4.7. A vertical cross-section (for its location see the label (a) in the Fig. 4.7 (b)) is shown in Fig. 4.8 where the cloud water content contours indicate that the cumulus touches the eastern flank of the stratospheric intrusion. The radiative cooling at the western upper edge of the cloud and the latent heat release inside the cloud induce a sharp heating gradient located precisely at the tropopause. This downward oriented heating gradient is responsible for the observed strong decrease of the PV values of near-tropopause air parcels, and hence for the pronounced STJ.

Furthermore, exchange occurs on the eastern flank of the streamer north of the cumulus cell, as can be seen in Fig. 4.7 (b). It appears to be located too far from the cloud cell to be induced by it directly. A west-east cross-section at 47°N is provided in Fig. 4.9. It shows that the cumulus is about 200 km further to the east. However, the strong cloud diabatic heating "pushes" isentropes downward and yields on the eastern flank of the intrusion a reduction of the static stability. This low static stability combined with the strong vertical shear due to the enhancement of the jet stream is likely to induce turbulence at the tropopause.
4.3. ANALYSIS OF PHYSICAL PROCESSES

Figure 4.8: Vertical west-east section across the stratospheric intrusion and the cumulus cloud cell by Sept 1 14UTC (see label (a) on the right panel of Fig. 4.7). The grey shading represents the cloud water content \(10^{-5}\) kg/kg\] (with highlighted contours at \(1 \times 10^{-5}\) kg/kg (dashed) and at 2, 4, 6 and 8 \(10^{-5}\) kg/kg (solid)). The bold solid lines are the 2 and 3PVU iso-lines. Light lines are the potential temperature iso-lines from 310 K to 340 K every 1 K. And the white lines represent the 10% (dash line) and 50% (solid line) partial cloud cover contours.

level. Richardson numbers are particularly small in this region, which further indicates the potential for turbulence in this region. The fact that contours of low diagnosed Richardson number do not reach the tropopause is not in contradiction with our hypothesis, but is interpreted as a consequence of the scale on which turbulence takes place (which is not accurately resolved in the numerical computation of the Richardson number). This kind of turbulence associated with large wind shear in the absence of clouds (referred to as clear air turbulence, CAT) has been observed by in-situ aircraft measurements (Shapiro 1976, Shapiro 1978) and implications for stratosphere-troposphere exchange have been discussed by Shapiro (1980). Finally, it is interesting to note how these turbulent processes act to extend the tropopause erosion, initiated by the proximity of the cloud cell near 43°N, further northward in a fairly continuous way.

The mixing of the tip is simply the consequence of its isolation caused by the strong diabatic processes related to the cloud cell.

4.3.2 The Cut-off Decay Episode

The decay of the cut-off went along with a splitting of the lower part of the stratospheric intrusion into a dynamically isolated blob of high potential vorticity (see Fig. 4.5). This splitting is a particularly interesting feature due to the localised occurrence of non-conservative
Figure 4.9: Vertical west-east section across the stratospheric intrusion on Sept 1 14UTC (see label (b) on the right panel of Fig. 4.7). The grey shading represents the horizontal velocity \( \text{[m s}^{-1}] \). The bold solid lines are the 2 and 3PVU iso-lines. Light lines are the potential temperature iso-lines from 310K to 340K every 1K. In addition are shown: the bold dash line is the 1 \( 10^{-5} \text{ kg/kg} \) contour of cloud water content; the white lines are the 10% (dash line) and 50% (solid line) partial cloud cover contours; and the closed solid lines are the Richardson number isolines 1, 1.5 and 2.

Processes. The analysis provided below shows that this splitting is linked to the radiatively induced vertical heating gradient (and subsequent negative potential vorticity rate) caused by the injection of humid air into the cut-off by turbulent mixing near the core of the jet stream.

Figures 4.10 (a) and (b) show the relative humidity on the 330 K isentrope at 04UTC and 09UTC of Sept 3, respectively. A wind speed maximum is found on the western flank of the cut-off. A tongue of moist air is advected cyclonically and quasi-isentropically southwards on the western side of the cut-off. The arrival of this tongue within the jet streak coincides with the appearance of a blob of humid air inside the cut-off (Fig. 4.10 (b)). The vertical section (Fig. 4.10 (c)) across this blob shows a zone of high relative humidity (50%) on the western side of the jet stream extending almost into the jet core. The location and shape of the blob of high relative humidity air found inside the cut-off suggests that humid air is injected into the cut-off via turbulent mixing near the jet centre. This precursor troposphere-to-stratosphere exchange can also be seen on the specific humidity evolution (not shown) and the low absolute amount of injected water vapour underlines the weakness of this exchange process. The humid air tongue west of the cut-off and the humid blob inside the cut-off are not associated with clouds.\(^7\)

\(^7\)The total relative humidity is smaller than 100% and consequently no clouds are produced in the model. An "ice relative humidity", evaluated with respect to ice, would give increasing values between 60% at
Figure 4.10: Advection of humidity on the 330 K isentrope along the jet stream and injection of water vapour into the cut-off. (a) and (b): the shaded field is the total relative humidity at 330 K and the contours for 30, 40 and 50% are highlighted. The bold contour denotes the 2PVU tropopause at 330 K. Solid contours show the horizontal velocity field (contours at 30, 32.5, 35, 37.5 and 40 ms\(^{-1}\)): (a) at 4UTC; and (b) at 9UTC. (c): cross-section at 42°N at 9UTC (corresponding to the line in panel (b)). Additionally are given the 3PVU isoline (bold line), the potential temperature isolines every 1 K (thin lines), and the partial cloud cover contours (white bold lines) at 10% (dashed) and 50% (solid).
The progressive intensification of the humid tropospheric filament inside the cut-off at 330 K is illustrated in Fig. 4.11, left panels at 10, 13 and 16 UTC of the same day. The slow erosion of the 2 PVU contour in the region where humid air is subject to enter the cut-off confirms the bidirectional nature of the exchange process, in harmony with the hypothesis of turbulent mixing. The right panels of Fig. 4.11 represent the evolution at a level located 2 K below (328 K). Here, the 2 PVU contour experiences a very different behaviour. It is “pushed” by the moist filament without any indication of inward mixing of moisture into the stratospheric sector. The difference of the dynamical evolutions of the 2 PVU contours between 330 K and 328 K is illuminating: it suggests that the cut-off splitting which occurs at 328 K is not due to turbulent mixing.

Fig. 4.12 (a) shows clearly the link between the moist filament and the local negative PV anomaly. The strong but localised negative PV anomaly is a signature of the presence of negative material PV rate in the lower half of the moist filament. Such a material PV rate is likely to be induced there by the negative vertical gradient of heating (see (4.1)) which is produced by the radiative effects in the moist filament: short-wave radiative cooling at the top, and long-wave radiative heating near the bottom (see for instance Manabe and Strickler (1964)). Note also the presence of a partial cloud cover within the filament (Fig. 4.12 (a)). This feature appears at 13 UTC (15%) and reaches a value of 45% at 16 UTC, which indicates that radiative processes in the filament are progressively strengthened after 13 UTC. However, the absence of partial cloud cover before 13 UTC suggests that radiative processes in pure water vapour were sufficient for the initiation of the cut-off splitting.

Thus, this (weak) vertical heating gradient together with the very high vertical vorticity inside the cut-off induce a very localised material reduction of potential vorticity, which in turn enables the mixing of stratospheric air within the troposphere.

This hypothesis is further confirmed by the analysis of the related trajectories. Figure 4.12 (b) shows a frequency distribution of the temporal evolution of the potential temperature along trajectories which undergo a stratosphere-to-troposphere exchange in the cut-off at 13 UTC. Exchange occurs within three well delimited potential temperature layers, with the most favourable one near 328 K. The computation of the material rate of potential temperature and potential vorticity along trajectories shows these layers to be dynamically different: trajectories of the highest layer (332 - 334 K) experience diabatic cooling (averaged value of $\dot{\theta} = -0.12 \text{ K/h}$) and a moderate PV loss ($\dot{Q} = -0.12 \text{ PVU/h}$), while in the middle layer (328 - 330 K), they are diabatically heated ($\dot{\theta} = 0.1 \text{ K/h}$) and have a stronger negative PV rate ($\dot{Q} = -0.2 \text{ PVU/h}$), and finally in the lowest layer (325 - 327 K) they have only very weak $\theta$ and PV rates. This layering is fully consistent with our hypothesis if we assume the cooling maximum to be placed at the top of the filament, between 330 and 332 K, and a heating maximum located at the bottom, near 327 K. The cut-off splitting is consequently realized mainly by heated parcels between 328 and 330 K.

This analysis clearly shows that presence of water vapour in the stratosphere can trigger significant stratosphere-to-troposphere exchange and influence the evolution of a tropopause.

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8 In the model, these two levels correspond to two different consecutive model levels.
Figure 4.11: Injection of the water vapour into the cut-off and induction of a negative PV anomaly below. The shaded field is the total relative humidity and contours for 30, 40 and 50% are highlighted. The bold closed contour is the 2PVU tropopause. Left panels on 330 K, and right on 328 K. (a) at 10UTC, (b) at 13UTC, (c) at 16UTC.
4.4 Variability with the PV-level

Analysis of fine-scale tropopause structures has shed light on the physical mechanisms which are presumably responsible for diagnosed exchanges. Most of the evidenced physical mechanisms were related to non-conservative processes originating in the troposphere.

In this section, an analysis of the variability of exchange with the considered iso-PV surface is performed with regard to the driving physical processes. The “upward influence” of the non-conservative processes and their signature on the exchange is discussed. The focus is on “downward” exchange\(^9\).

Figure 4.13 shows the time evolution of the mass exchange estimates at various PV-levels between 1.5 and 5PVU for both flux directions. The sensitivity is large and marked by strong temporal variations indicating a dependence on the exchange mechanism. The break-up of the streamer and the cut-off decay (as discussed before for the 2PVU tropopause) lead to downward exchange which is highly dependent on the PV-level. The exchange intensity monotonically decreases with the PV-level, indicating a progressive reduction of the effects of the tropospheric non-conservative processes. However, this sensitivity seems to

\(^9\)“Downward (upward) exchange” is used here for exchange towards lower (higher) PV values.
Figure 4.13: Temporal evolution of the 12 h accumulated mass exchange at various PV levels. Left: stratosphere-to-troposphere exchange, right: troposphere-to-stratosphere exchange. Time relative to Sept 1 00UTC. Values are drawn at the middle of the 12h integration periods. Unit: $10^9$ kg s$^{-1}$. The investigation is performed over the reduced domain: 10W-20E, 38N-58N.

be confined to the levels below 4PVU, and the temporal evolution of exchange at this level and above reveals a slightly different pattern. This can also be seen (to a smaller extent) in the troposphere-to-stratosphere exchange fluxes.

Figure 4.14 details the exchange estimates across each PV-level for the streamer break-up period (12 - 24 h in Fig. 4.13). The exchange pattern at the streamer’s tip appears to be highly sensitive to the PV-level. In contrast on the streamer’s flank, similar exchange patterns are present at each level, but with decreasing intensities towards higher levels. This indicates that the diabatic and associated turbulent processes at the flank induce exchange over a significant vertical depth and are “penetrative” processes. On the other hand, a local exchange maximum occurs near the streamer’s tip on levels below 2.5PVU at a location which depends on the level. This local maximum corresponds to the mixing of the streamer’s tip, and as can be seen on temporal evolutions (not shown), the mixing of various PV iso-surfaces is temporally differentiated: the 2.5PVU surface is mixed at 12UTC, the 2.0PVU at 14UTC, while the 1.5PVU is finally mixed at 23UTC. In effect the advection of the streamer during this time period accounts for the geographical variability of the exchange maximum. Above 2.5PVU, the streamer evolution is slightly different in the sense that the tip is less developed as its erosion took place earlier in the vicinity of the cloud cell. This suggests that the mixing of the tip starts indeed near the zone of strong diabatic processes associated with the cloud cell at high PV-levels and then “propagates” in the tip towards lower PV-levels along with the flow motion.

The variability of downward exchange with the considered level is also large in the cut-off...
Figure 4.14: Exchanged mass at various PV levels for the period Sept 1 12-24UTC in $10^0 [\text{kg/s}]$. Only stratosphere-to-troposphere fluxes are represented. Bold lines represent 325 and 330 K contours at the corresponding PV level at 18UTC.
decay phase (see time period for $t \geq 36\,\text{h}$ in Fig. 4.13) and can be interpreted in term of the specific physical processes responsible for the exchange. The negative PV tendency induced by the humid filament injected into the cut-off is located below a strong vertical gradient in potential vorticity. The filament has consequently an action on the PV-levels which are present under this large gradient zone. Temporal evolutions of the various PV iso-surfaces show that the 3.5PVU iso-surface is the uppermost where an erosion similar to the one described at 2PVU still occurs, but to a smaller extent (in agreement with Fig. 4.13).

More globally, this particular case study reveals two regimes in the exchange behaviour. Below 4PVU, iso-PV surfaces possess very fine-scale structures in relation to the tropospheric non-conservative sources and the exchange patterns are strongly shaped by these sources. However, both the geographical distributions and the intensities of exchange vary significantly with the considered PV-level. Above 4PVU, iso-PV surfaces are much smoother and the exchange is mostly due to the tropospheric non-conservative sources which are penetrative enough. Geographical distributions and intensities of exchange show a reduced variability with the considered level. This indicates that exchange across the 4PVU iso-surface is likely to be easier to estimate and to interpret. However, this does not argue for a choice of a tropopause at 4PVU. Instead, the discrepancies in the location of exchange across the 4PVU and 2PVU iso-surfaces, in particular in the vertical, coupled with the chemical properties of the 2 - 4PVU layer (as revealed for instance by the PV-ozone correlations, Brunner (1998)), suggest that a low enough iso-PV surface (in particular 2PVU) can be necessary to represent correctly the stratospheric inflow into the troposphere whenever three-dimensional forcing issues are addressed.

### 4.5 Effects of the Residence Time Criterion

The residence time criterion was introduced to eliminate spurious events due to numerical errors. In Fig. 4.15 the time evolution of the horizontally integrated mass fluxes are compared when estimated (a) without residence time criterion and (b) with $\tau_{\text{crit}} = 12\,\text{h}$. Without the criterion, both upward and downward fluxes are larger by a factor of 4 to 5, while the net fluxes are very similar in the two cases. In effect numerous events are removed using the residence time criterion without altering the net flux, and hence the removed events are mostly associated with oscillating trajectories (see section 3.2). Thus, although (Lagrangian) estimates of exchange mass fluxes without an additional criterion can be dominated by numerical effects, the Lagrangian approach also offers a means of defining a reliable criterion that seems to remove "artificial" events. Real exchanges with short residence times are likely to represent a (small) part of the removed events, but it is in general difficult to quantify their relative amount, and they are assumed to play a weak role in the chemical/radiative forcing due to their short interaction time.

STE fluxes estimated with different critical residence times ($\tau_{\text{crit}} = 12, 24, 36$ and 48 h) are presented in Fig. 4.16. If these estimates are assumed to be free of numerical errors, then their variability with the critical residence time is related to the residence of parcels within the troposphere and stratosphere. The convergence of the curves at high critical residence times indicates a decreasing sensitivity towards large values of $\tau_{\text{crit}}$, and thus a significant
Figure 4.15: Time evolution of the horizontally integrated mass exchange estimated with and without the critical residence time $\tau_{\text{crit}} = 12\text{h}$. Left: stratosphere-to-troposphere exchange, centre: troposphere-to-stratosphere exchange, right: net stratosphere-to-troposphere exchange. Time is relative to Sept 1 00UTC. Values are drawn at the centre of the 12h integration periods. Unit: $10^9$ kg s$^{-1}$. Integration domain: 10W-20E, 38N-58N.

part of the exchange events have a residence time longer than a week. Moreover, the small effect of the residence time on the estimated net fluxes suggests that the net exchange is mostly realized by long residing parcels.

4.6 Conclusion and Further Remarks

The method developed in the previous Chapter has been applied to one particular synoptic event involving a baroclinic wave breaking and the subsequent cut-off decay.

The total estimated cross-tropopause mass fluxes fall in the range of other estimates of synoptic scale developments available from the literature (see Table 2.2 in the previous Chapter) with $S\rightarrow T$ fluxes of $2.3 \times 10^9$ kg s$^{-1}$ and $T\rightarrow S$ fluxes of $1.6 \times 10^9$ kg s$^{-1}$. A three-dimensional perspective has allowed to consider the occurrence of exchange in relation to tropopause mesoscale structures. This has enabled a qualitative validation of the results, and has evidenced subtle exchange mechanisms.

In this case study, strong exchange occurred in the explosive streamer break-up and in the cut-off decay. Exchange occurring during the streamer break-up episode has been shown to relate to diabatic and turbulent effects at the tropopause level due to a strong cloud cell. The cut-off decay episode has been associated with the splitting of the cut-off within a thin isentropic layer, which is caused by the injection of a moist filament into the cut-off and the related radiative processes.

The variability of exchange with the considered iso-PV level has been explored, and two different regimes have been found: the exchange flux and its geographical distribution varies significantly with the PV-level below 4PVU, while above the sensitivity is reduced.

The sensitivity to the critical residence time has shown that a significant part of the ex-
4.6. CONCLUSION AND FURTHER REMARKS

change flux is associated with residence times smaller than two days, and that "irreversible" exchange (residence time much larger than two days) determines the net exchange flux in this case study.

The sensitivity of the exchange estimates to the data resolution has been analysed and the robustness of the method has been explored (see Appendix C). The sensitivity is found to be relatively low for the range of resolutions emphasised here, and the causes evidenced by the analysis are shown to be dominantly related to the scale of physical processes or to inconsistencies in the data itself. As a consequence, reliable estimates can be expected from consistent data sets with resolutions down to (1.0°, 6h), as for instance with ECMWF (re)analysis data.

Figure 4.16: Same as Fig. 4.15, but for different values of the critical residence time: $\tau_{\text{crit}} = 12, 24, 36$ and $48$ h.
Chapter 5

A One-Year Climatology of STE

The exchange of mass between the stratosphere and the troposphere forces both the strato-
sphere and the troposphere. Chapter 4 has shown that large amounts of stratospheric mass
can be injected into the troposphere during a synoptic development like a baroclinic wave
breaking. The effects that such exchange episodes induce on the large scale are important for
number of purposes from the synoptic-scale to the large-scale. A precise knowledge and un-
derstanding of STE is therefore required also on the global scale. Our Lagrangian approach
for the diagnosis of STE has been extensively discussed and checked on the synoptic-scale
in the Chapters 3 and 4, and is applied in this and the next Chapters to a one-year north-
hemispheric climatology with the cutting-edge potential of providing a global coverage while
resolving dynamical and physical processes on the synoptic-scale (cf. Chapter 2). The ability
of the method to realistically reproduce the large-scale signatures that have been detected
from global methods is checked quantitatively and qualitatively throughout the next Chap-
ters.

The basic results of the climatology are dressed in the present Chapter. The application
of the method is firstly presented. Then the zonal mean and the geographical distribution
of exchange fluxes are discussed. The vertical location of the exchange is also described.
Finally, the part of the exchange fluxes which interact with the boundary layer is examined.

5.1 Methodology

The Lagrangian approach defined in Chapter 3 is applied here to provide a one-year cli-
tatology of STE for the period May 1995 to April 1996.\(^1\)

The set of trajectories that represents the atmospheric flow motion has been computed
using the six-hourly ECMWF T213L31 analysis data, interpolated on a \(1^\circ \times 1^\circ\) longi-
tude/latitude grid. Secondary variables like potential temperature and PV have been calcu-
lated on the hybrid model levels. The parameters of the method (cf. Chapter 3) have been
chosen consistently with the data resolution and the climatological nature of the accounted

\(^1\)This period has been chosen due to the availability of ozone measurements from a commercial airliner
(project NOXAR, Brunner (1998)) which has be used to estimate the cross-tropopause flux of ozone in the
North Atlantic region (see Chapter 8).
estimates. The trajectory starting grid is such that the elementary time period $T$ is 24 h; the whole northern hemisphere is covered with a vertical extent from 590 to 50 hPa; and the starting grid resolution is 80 km horizontally and 30 hPa vertically. Exchange events are identified via the model defined in Fig. 3.2 with a critical residence time $\tau_{\text{crit}} = 48 \text{ h}$ to give the emphasis on the exchange events which have a significant interaction time. The sensitivity of the exchange mass flux with the critical residence time is discussed in the next Chapter.

It should be pointed out that the results of this one-year climatology are subject to the interannual variability.

### 5.2 The Zonal Mean Picture of STE

#### 5.2.1 Separated $S \rightarrow T$ and $T \rightarrow S$ Exchange Fluxes

Figure 5.1 illustrates the zonally integrated $S \rightarrow T$ and $T \rightarrow S$ exchange mass fluxes on the northern hemisphere for the months from May 1995 to April 1996 (hereafter the notations $S \rightarrow T$ and $T \rightarrow S$ are used for stratosphere-to-troposphere and troposphere-to-stratosphere exchange, respectively). $S \rightarrow T$ exchange is most intense in mid-latitudes between 30 and 60°N, whereas $T \rightarrow S$ mass flux shows a maximum in the subtropics (15-30°N), comparatively low values from 30-40°N, and again larger values in the northern mid-latitudes between 45 and 60°N. Generally the cross-tropopause mass fluxes are largest in winter and smallest in summer.

Another representation of the exchange flux which is significantly different from the zonally integration is the zonally averaged mass flux. While the former provides insight into the meridional distribution of the vertical flux, the latter shows more directly the "exchange

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2 The resulting number of trajectories is $\sim 2.6 \times 10^6$ for the whole year.

3 A 15-years climatology is being presently performed on the basis of the method and results presented here, and will appear in a future publication.
Figure 5.2: Same as Fig. 5.1 but for zonally averaged exchange mass fluxes. Unit is in kg km$^{-2}$ s$^{-1}$.

activity”. Figure 5.2 gives the spatio-temporal structure of the zonal mean exchange fluxes. The decrease of the fluxes north of 70° present in the zonally integrated fluxes has disappeared. The T→S exchange activity increases north of 40°N towards the pole for all seasons. The dominance of the low latitude band of maximum T→S exchange upon the high latitude one in the zonally integrated flux (cf. Fig. 5.1) is therefore due to the length of zonal bands.

5.2.2 Net and Two-Way Exchange Fluxes

The S→T and T→S exchange mass fluxes ($F^{S\rightarrow T}$ and $F^{T\rightarrow S}$) can be recombined in two different fluxes which contain together the same information as the original fluxes:

- the net exchange flux ($F^{\text{net}}(x, y, t) = F^{S\rightarrow T}(x, y, t) - F^{T\rightarrow S}(x, y, t)$);

- the symmetric two-way exchange flux which represents the common part of the S→T and T→S exchange fluxes which cancels in the net exchange flux ($F^{\text{2way}}(x, y, t) = \min\{F^{S\rightarrow T}(x, y, t); F^{T\rightarrow S}(x, y, t)\}$)\(^4\).

Figure 5.3 shows the zonally integrated net exchange flux. The net exchange flux is upward south of 30°N, downward between 30 and 65°N, and weakly upward north of 65°N. The seasonal cycle is different in the two belts of upward and downward fluxes: the tropical belt has no marked cycle, and the midlatitude belt has a clear maximum in winter-spring and minimum in summer.

The zonally integrated two-way exchange distribution reveals a meridional pattern with a maximum in the midlatitudes near 50°N (cf. Fig. 5.4). In contrast to the net exchange the seasonal variability of two-way exchange is weak with slightly enhanced values during winter and reduced values during summer. The zonally averaged two-way exchange flux has a different meridional profile: a zone of uniformly large two-way exchange “activity” is found

\(^4\)The term “two-way exchange” is used here in a large-scale meaning and do not imply any material link between individual upward and downward exchanges.
5.3 The Geographical Distribution of STE

Figures 5.5 and 5.6 show the geographical distributions of the S→T and T→S exchange mass fluxes together with a measure for the dynamical storm-track activity\(^5\) on the northern hemisphere for the four seasons.

Maxima of S→T exchange (Fig. 5.5) occur within the midlatitude storm-track regions over the Pacific and Atlantic oceans, except for summer, where the zonal variability is very weak. Within the storm-tracks S→T exchange shows a lot of variability and there is no general preference for S→T exchange to occur either in the entrance or exit regions. Another area with enhanced S→T exchange activity are the Asian mountains during winter.

\(^5\)The 500hPa block-filtered transient eddy geopotential height field (Hoskins et al. 1989, Hall and Plumb 1994) is used as a measure for the dynamic storm-track.
and spring. Other preferred continental regions for $S\rightarrow T$ exchange throughout the year are western North America and the area to the west/south-west of the Alps.

$T\rightarrow S$ exchange on the other hand (Fig. 5.6) has pronounced maxima in the subtropics and in the northern parts of the mid-latitude storm-tracks. Within the subtropical belt there is substantial seasonal variability with maxima over the Atlantic in winter, the Pacific in spring, and south-eastern Asia during the Monsoon season (summer/autumn). Notable regions with enhanced $T\rightarrow S$ exchange activity in the midlatitudes are eastern Canada during all seasons, and the (eastern) Mediterranean in summer and autumn.

5.4 The Vertical Location of Exchange

The vertical distribution of the exchange locations are shown for the winter and summer seasons for $S\rightarrow T$ exchange and $T\rightarrow S$ exchange in Fig. 5.7.

In the extra-tropics $S\rightarrow T$ exchange occurs typically some 0-200 hPa below the height of the climatological tropopause. This indicates that $S\rightarrow T$ exchange is frequently associated with tropopause folds which come down as low as 600 hPa in the region from 30 - 50°N during winter. In the summer season only very few exchange events happen below 500 hPa.

In contrast to $S\rightarrow T$ exchange, midlatitude $T\rightarrow S$ exchange occurs typically some 50 hPa above the climatological tropopause height in all seasons. The subtropical events take place at a fairly constant level of about 100 hPa. It is notable that only little exchange ($S\rightarrow T$ and $T\rightarrow S$) takes place in the steepest climatologic tropopause region which occurs near 20 - 30°N during winter.

During spring and autumn the vertical distributions (not shown) are intermediate to the ones presented for the winter and summer seasons.

5.5 The Deep Exchange Events

Finally we focus on vertically "deep exchange events", that is on $S\rightarrow T$ exchange events where the originally stratospheric air penetrates to levels below 700 hPa, and on $T\rightarrow S$ exchange events where originally tropospheric air from below the 700 hPa level is injected into the stratosphere within a couple of days. These events have potentially a particularly important chemical impact as they lead to the interaction of (near) boundary layer and stratospheric air, which are characterised by very different chemical compositions.

Figure 5.8 shows the zonally integrated cross-tropopause mass fluxes associated with deep $S\rightarrow T$ and $T\rightarrow S$ exchange events for the months from May 1995 to April 1996. Deep $S\rightarrow T$ exchange is characterised by a marked seasonal cycle (much more prominent than when considering all exchange events, see Fig. 5.1) with a maximum in winter near 45°N and a reduction by a factor of ten during summer. Integrated over the hemisphere, deep events account for about 5% of the total $S\rightarrow T$ mass flux during winter. For deep $T\rightarrow S$ exchange events the seasonal variability is much weaker, although they also occur most frequently during the cold season. Comparison with Fig. 5.1 reveals that a substantial part of the northern midlatitude $T\rightarrow S$ exchange events are deep, but almost none of the subtropical ones.
Figure 5.5: Geographical distribution of the S→T exchange mass flux for the four seasons. Values are in kg km$^{-2}$ s$^{-1}$. a) winter (DJF), b) spring (MAM), c) summer (JJA) and d) autumn (SON). Overlayed are two contours for the 500 hPa high-pass filtered transient eddy geopotential height field as a measure for the storm-track activity (solid line for 50 m, and dash-dotted line for 70 m).
5.5. THE DEEP EXCHANGE EVENTS

Figure 5.6: Same as Fig. 5.5 but for the T→S exchange mass flux.
Figure 5.7: Vertical-meridional distribution of the location of the S→T (left panels) and T→S (right panels) exchange for winter (upper panels) and summer (bottom panels). Here a threshold residence time of 96 hours has been used. Values denote the number of events within a $3^\circ \times 20\text{hPa}$ large grid box.
Figure 5.8: Zonally integrated exchange mass flux associated with deep exchange in $10^6$ kg s$^{-1}$ per 1 km broad zonal band. Left: S$\rightarrow$T exchange; right: T$\rightarrow$S exchange. X-axis is the month, starting in May 1995 and ending in April 1996; and y-axis is the latitude.

The geographical distribution of deep exchange events is illustrated in Fig. 5.9 for the winter season. During the winter deep S$\rightarrow$T and T$\rightarrow$S exchange almost exclusively occur within the regions of the dynamical storm-tracks, and in the Arctic region for deep T$\rightarrow$S exchange. Deep S$\rightarrow$T exchange has distinct maxima in the preferred areas for cyclogenesis near the Asian and American east coasts, and in the southern parts of the storm-track exits over California and the Mediterranean region. For spring and autumn the qualitatively good general correlation with the storm-track regions persists although the location of the local maxima changes (not shown). During summer a few deep S$\rightarrow$T exchange events occur over the American west coast and Scandinavia, whereas deep T$\rightarrow$S exchange is localised in the central North Pacific and near Greenland (not shown).

Figure 5.10 indicates, again for the winter season only, where deep S$\rightarrow$T exchange air parcels can be observed below the 700 hPa during the four days after the exchange, and from what locations below the same level the air is rapidly transported into the stratosphere during the following days. These diagrams provide important information as they point to the regions where for instance stratospheric ozone intrusions can be observed near the ground (deep S$\rightarrow$T exchange), or from where anthropogenic pollutants are likely to be conveyed into the stratosphere (deep T$\rightarrow$S exchange). Again for the considered winter stratospheric intrusion preferentially occur along the southern border of the storm-tracks and in continental areas to the south of their exit regions (Baja California, Mediterranean and Northern Africa). On the other hand, the low points associated with deep TSE events reveal two prominent hot spots in the south-western parts of the Atlantic and Pacific storm-tracks and almost zero values over the Eurasian continent.
Figure 5.9: Exchange mass flux associated to deep S→T (left) and T→S (right) exchange events for winter. Values are in kg km\(^{-2}\) s\(^{-1}\). Overlayed are two contours for the 500 hPa high-pass filtered transient eddy geopotential height field as a measure for the storm-track activity (solid line for 50 m, and dash-dotted line for 70 m).

Figure 5.10: Geographical distribution of the trajectory points below 700 hPa during winter associated with deep exchange events, for S→T (left) and T→S (right) exchange events. Values correspond to the total number of deep exchange trajectory points below 700 hPa within a 3° × 3° grid box. Overlayed are two contours for the 500 hPa high-pass filtered transient eddy geopotential height field as a measure for the storm-track activity (solid line for 50 m, and dash-dotted line for 70 m).
5.6 Discussion and Further Remarks

The application of the Lagrangian method to the ECMWF data set implies some limitations. First, as already mentioned, the estimates are subject to several limitations in the tropics, and therefore this study focusses on the extra-tropical region north of 10° - 15°N and is not intended to quantify the upward mass flux across the tropical tropopause. Secondly, the deep convection is likely to play a significant role during summer in the extra-tropics, but is not explicitly resolved by the ECMWF model. Hence, our deep exchange estimates might be underestimated in summer. However, this effect is likely to be attenuated since the influences of cumulus clouds on the atmospheric stability and hence on the tropopause height are captured (in regions where vertical temperature soundings are available).

This one-year climatology has revealed novel and important characteristics of stratosphere-troposphere exchange in the extra-tropics. Detailed information on the horizontal and vertical location of exchange has been provided. A marked zonal asymmetry has been shown, with the Atlantic and Pacific storm-tracks as preferred regions for cross-tropopause transport, especially during the cold season. In the vertical S→T (T→S) exchange occurs typically in a 150 hPa (80 hPa) layer below (above) the climatological height of the tropopause. This indicates that S→T exchange takes place preferentially within transient systems with a low tropopause, like for instance tropopause folds (conversely for T→S exchange).

A two-way exchange flux has been derived in parallel to the net exchange flux and has been shown to be of equal importance of the latter, but is characterised by a different meridional distribution and only a weak annual cycle. This implicates that a knowledge limited to the net flux is incomplete for purposes related to the radiative and chemical forcing induced by STE.

Additionally, substantial vertical transport associated with cross-tropopause exchange has been detected, especially during winter. Of particular chemical importance are vertically "deep exchange events" where air is transported from the stratosphere to the atmospheric boundary layer (or vice versa) within a couple of days. Geographical distributions of the presence of deeply exchanged parcels below 700 hPa have highlighted (i) the regions where ozone-rich air from the stratosphere is likely to influence the boundary layer, and (ii) the regions of the boundary layer from where natural and anthropogenic gases are likely to attain rapidly the stratosphere. In particular, emissions from the near Japan and the American east coast enter much more frequently the stratosphere than emissions from other areas including Europe.

These estimates of STE are in reasonable agreement with estimates available from the literature (cf. section 2.2 for a review). The seasonal cycle is comparable to the one of global flux evaluations at higher levels. As mentioned in section 2.2, Appenzeller et al. (1996b) estimated the net flux across the extra-tropical tropopause by evaluating the flux across the

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6 The limitations in the tropics are related to (i) the lack of southern hemispheric data important for the proper computation of trajectories near the equator; and (ii) the generally lower quality of the analysis data in this region, due to the poor spatial coverage of measurements and the importance of deep convection which is not explicitly resolved by the ECMWF model.

7 These regions are related to the storm-tracks and therefore might have small interannual variability.
380 K isentrope and estimating the lowermost stratosphere mass variation. Fig. 2.3 (section 2.2) represents the annual cycle of their estimates. Note that this flux corresponds to the integration of the cross-tropopause flux north of the latitude where the two iso-surfaces 2PVU and 380 K intersect. Fig. 5.11 shows the annual cycle of our estimated flux, integrated north to 18°, 24°, 30° and 38°N. Provided the interannual variability, the differences in the methods, and the different southern latitudes of the integration region, the comparison of these two temporal evolutions show very good agreement: the seasonal cycles are very similar, with a maximum in winter - spring and a minimum in late summer - autumn; and the magnitudes are similar, with values of Appenzeller et al. (1996b) varying between 5 and 15 $10^9$ kg s$^{-1}$ and our between 0 and 15 $10^9$ kg s$^{-1}$, depending on the considered southern integration boundary.

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8The lowermost stratosphere is embedded between the 2PVU tropopause and the 380 K isentrope.
Chapter 6

The Residence Time of Exchange

In the preceding Chapter, a one-year climatology of STE has been presented on the northern hemisphere. The estimates were based upon a critical residence time $\tau_{\text{crit}} = 48\,\text{h}$, in order to emphasise the exchange events having a significant interaction time within the stratosphere and the troposphere.

However, for purposes which are related to the interaction time of exchanged parcels, like the chemical and radiative forcings induced by STE, the entire (statistical) distribution of the residence time of the exchange flux can play a role. In effect, depending on the time-scale of the residence times and of the chemical/radiative processes, the actual forcing induced by STE can be closely related to a ratio of residence time and chemical/radiative time-scales.

This effect can have important implications for the parameterisation of STE in coupled GCM-chemistry models. It is clear that if such a global model can not resolve the exchange processes, it will not reproduce reasonable residence time distributions as well. Thus, the additional parameterisation of the residence time distribution of exchange might also be necessary.

The thrust of this Chapter is to quantify the sensitivity of the estimated exchange mass fluxes $F^{S\rightarrow T}$ and $F^{T\rightarrow S}$ to the critical residence time $\tau_{\text{crit}}$, and its spatio-temporal variability.\footnote{The issue of assessing the actual chemical impact of STE using the realistic residence time distribution evaluated here is addressed in Chapter 9.} The method for estimating STE follows Chapter 5, but with different imposed values of $\tau_{\text{crit}}$, ranging from 24, 36,... to 96 h. The Chapter contains firstly the analysis of the dependence of exchange mass on the critical residence time. Then the residence time distribution is identified and its properties are discussed. Finally, the results are discussed with regard to their spatio-temporal variability.

6.1 Sensitivity of Exchange Fluxes to $\tau_{\text{crit}}$

Figure 6.1 provides the annual cycle of the zonally integrated exchange mass flux for the period May 1995 - April 1996, for the values $\tau_{\text{crit}} = 24, 48, 72$ and 96 h. The sensitivity of the spatio-temporal patterns to $\tau_{\text{crit}}$ is different for the two exchange directions. The overall pattern of stratosphere-to-troposphere exchange estimates (meridional belt of maximum
exchange around 40°N with a maximum in January and a minimum in August) remains almost unchanged in the considered range of \( \tau_{\text{crit}} \). A marked sensitivity however appears in the tropical region 10° - 30°N: the stratosphere-to-troposphere exchange flux decreases strongly in this region as \( \tau_{\text{crit}} \) increases, which has the effect to enhance the meridional gradient at the southern edge (30° - 40°N) of the main exchange belt.

The general spatio-temporal pattern of the troposphere-to-stratosphere exchange flux is composed of two meridional belts of exchange (10° - 40°N and 40° - 60°N) for every \( \tau_{\text{crit}} \). But their relative importance changes significantly with \( \tau_{\text{crit}} \): for \( \tau_{\text{crit}} = 24 \) h, the subtropical exchange belt dominates, and for \( \tau_{\text{crit}} = 96 \) h, the two exchange belts have similar amplitudes. The annual variability within the two meridional belts is different. The midlatitude belt has a similar annual cycle for all values of \( \tau_{\text{crit}} \) (maximum in January and minimum in July). In the subtropical belt, the only distinct feature for \( \tau_{\text{crit}} > 24 \) h is a minimum in December which is more pronounced as \( \tau_{\text{crit}} \) is increased.

In addition, a strong decrease of the overall flux amplitude with \( \tau_{\text{crit}} \) is visible from the scales in Fig. 6.1. Together with the spatio-temporal variability of the exchange flux with \( \tau_{\text{crit}} \) this demonstrates that the dependence of the exchange flux on the critical residence time is (i) strong and (ii) spatially and temporally differentiated. Therefore, the distribution of residence time associated with the exchange fluxes is likely to play a significant role when assessing the impact of STE.

6.2 Analysis of the Distribution of Residence Time

The annual and hemispheric\(^2\) mean exchange mass flux is represented in Fig. 6.2 as a function of the critical residence time \( \tau_{\text{crit}} \) for stratosphere-to-troposphere (S\(\rightarrow\)T, crosses) and troposphere-to-stratosphere (T\(\rightarrow\)S, diamonds) exchange. Note that the two exchange directions experience a similar rapid decay with \( \tau_{\text{crit}} \): the flux corresponding to parcels with 1 day \( \leq \tau_{\text{crit}} \leq 4 \) days is nearly three times as large as the flux of parcels remaining exchanged for more than 4 days.

The variation of the mean fluxes with \( \tau_{\text{crit}} \) can be accurately\(^3\) approached by an exponential function:

\[
<F(\tau_{\text{crit}}; x, y, t) >_{x,y,t} \approx (\bar{F}_0 - \bar{F}_\infty) \exp \left( - \ln 2 \frac{\tau_{\text{crit}}}{\tilde{\tau}_{1/2}} \right) + \bar{F}_\infty
\]

(6.1)

where \( < \cdot >_{x,y,t} \) denotes the averaging over the year and the hemisphere, and overbars are used for the corresponding exponential fit parameters. \( \bar{F}_0 \) and \( \bar{F}_\infty \) are the extrapolations of the exponential fit for \( \tau_{\text{crit}} = 0 \) and \( \tau_{\text{crit}} \rightarrow \infty \), respectively. \( \tilde{\tau}_{1/2} \) represents the rate of decay of the exponential fit such that half of the decay takes place within a period of \( \tilde{\tau}_{1/2} \). The exponential functions which fit the estimated mass fluxes are shown in Fig. 6.2 and the corresponding parameters are given in Table 6.1. The three parameters \( \tilde{\tau}_{1/2} \), \( \bar{F}_0 \) and \( \bar{F}_\infty \) entirely determine the dependence of \( F \) on \( \tau_{\text{crit}} \) within the time window 24 - 96 h.

\(^2\)Here the calculations do not include the latitudes below 10°N.

\(^3\)The linear determination coefficient of the exponential fits are larger than 0.99 and the deviations are not significant for the five percent level of significance of the Chi-Square test.
Figure 6.1: Zonally integrated exchange mass fluxes (left: S→T, right: T→S) in $10^6$ kg s$^{-1}$ per 1 km broad zonal band. The x-axis denotes the month, starting in May 1995 and ending in April 1996; and the y-axis is the latitude. The critical residence times $\tau_{crit}$ are (a) 24 h; (b) 48 h; (c) 72 h; (d) 96 h.
CHAPTER 6. THE RESIDENCE TIME OF EXCHANGE

Figure 6.2: Hemispheric and annual mean exchange mass fluxes for several critical residence times $\tau_{\text{crit}}$. Crosses (diamonds) represent stratosphere-to-troposphere (troposphere-to-stratosphere) exchange fluxes. Solid line: exponential fit of the stratosphere-to-troposphere estimates; dotted line: exponential fit of the troposphere-to-stratosphere estimates. Fitting parameters are provided in Table 6.1 following the formulation (6.1).

<table>
<thead>
<tr>
<th>$\tau_{1/2}$ [h]</th>
<th>$F_0$ [kg km$^{-2}$ s$^{-1}$]</th>
<th>$F_\infty$ [kg km$^{-2}$ s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>S$\rightarrow$T</td>
<td>15.8</td>
<td>84</td>
</tr>
<tr>
<td>T$\rightarrow$S</td>
<td>15.7</td>
<td>61</td>
</tr>
</tbody>
</table>

Table 6.1: Values of the fit parameters for the dependence on $\tau_{\text{crit}}$ of the mean hemispheric and annual exchange fluxes. See Fig. 6.2 and the formulation (6.1).

This exponential dependence, shown above on the hemispheric and annual scale, also applies to the regional scale on monthly mean exchange estimates. Exponential fit of the dependence of estimates with $\tau_{\text{crit}}$ have been calculated independently for each grid-square ($3^\circ \times 3^\circ$) of monthly mean geographical distributions of exchange estimates. The goodness of fit is individually checked using the five percent level of significance of the Chi-Square test, and the rejected fits are excluded. Note that rejected fits are globally rare, but regions where their incidence may have some effect on the average are highlighted hereafter via the statistical significance of the averages.

The general exponential shape of the dependence of exchange fluxes with $\tau_{\text{crit}}$ is an intrinsic property of the exchange fluxes which can be quantified via the three parameters of the fit ($F_0$, $F_\infty$, $\tau_{1/2}$). Hence, these parameters provide direct insight into the Lagrangian properties of exchange. However, their interpretation should be limited to the available temporal window, as other regimes might take place for values of $\tau_{\text{crit}}$ between 0 and 24 h and larger than 96 h. Consistently, the extrapolation of “observed” values to $F_0$ for $\tau_{\text{crit}} = 0$ is considered here as not reasonable. But $F_\infty$ represents the irreversible part of the mass
6.3. GEOGRAPHICALLY RESOLVED ESTIMATES of $\tau_{1/2}$ and $F_\infty$

Figures 6.3 and 6.4 show the zonal mean characteristic residence times $\tau_{1/2}$ and irreversible mass fluxes $F_\infty$ as functions of the latitude and month for the two directions of exchange. Additionally are given the 90 and 95% levels of statistical significance of the fit parameters.

The values of $\tau_{1/2}$ show a large spatio-temporal variability (between 10 and 24 h). For the two flux directions, the characteristic residence time generally increase towards the north, and has a distinct seasonal cycle with the minimum in November-January. The stratosphere-to-troposphere exchange flux possesses an enhanced northward gradient of $\tau_{1/2}$ near 40°N which splits the hemisphere into a sector of low residence times with a minimum near 25°N, and a sector of larger values of $\tau_{1/2}$ in the north with a nearly homogeneous meridional distribution. A slightly different pattern is found for the troposphere-to-stratosphere exchange flux. A broad zone of minimum $\tau_{1/2}$ appears near 30°N, extending between 10 and 50°N. And north of 50°N, the characteristic residence time tends to increase toward the pole.

The spatio-temporal patterns of the irreversible part $F_\infty$ of the exchange mass flux are in line with the series of patterns provided in Fig. 6.1 for increasing $\tau_{crit}$, and are very similar to the fluxes for $\tau_{crit} = 96$ h, but with slightly smaller amplitudes (by a factor of 0.8 - 0.85).

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4A $\tau_{crit} = \tau_{1/2} + 24$ h corresponds to about half of the available decay.
6.4 Conclusion

The exchange mass fluxes reveal a strong sensitivity to the critical residence time $\tau_{\text{crit}}$ in the time window 24 - 96 h, both in the hemispheric and annual mean fluxes, and in the spatio-temporal patterns. This demonstrates that the exchange flux is not in general irreversible on this time window.

The mass flux experiences a rapid decrease with the critical residence time which can be accurately fitted by an exponential function, both on the $1^\circ \times 1^\circ$ bin/month and hemisphere/annual scale. The parameters of the fit provide two measures relevant for the residence time distribution: the characteristic residence time $\tau_{1/2}$ which gives an overall measure of the residence time distribution of the mass flux; and the "irreversible" component of the mass flux $F_\infty$ - the term "irreversible" is used here relative to the available time window to designate a period longer than one week. The characteristic residence times for both upward and downward exchange fluxes possess a distinct seasonal and meridional variability (a minimum in November - January and an overall northward gradient). Typical values between 10 and 14 h are found south of 40°N and between 18 and 22 h further north. The irreversible exchange fluxes are close to those with $\tau_{\text{crit}} = 96$ h.

The exponential shape of the dependence of exchange fluxes on $\tau_{\text{crit}}$ and the values of $\tau_{1/2}$, $F_0$ and $F_\infty$ are relevant for the assessment of the chemical (and radiative) impacts of STE. Depending on the temporal scale of the chemistry, the impact of mass exchange is modulated by the characteristic residence time, the irreversible mass flux or both. This issue is further analysed in Chapter 9.

This analysis clearly reveals that the information contained in mass flux estimates obtained with an Eulerian method (i.e. corresponding to $\tau_{\text{crit}} \approx 0$) does certainly not suffice for purposes which involve the interaction of exchanged parcels within the stratosphere and troposphere. Eulerian estimated flux can not be considered as irreversible.

The analysis of the residence time distribution of the exchange fluxes has shed light on a fundamental and intrinsic Lagrangian property of the stratosphere-troposphere exchange.
Chapter 7

Generalised Cross - iso PV Transport Near the Tropopause

The Lagrangian method developed in this study has been applied so far to the 2PVU tropopause and results on the synoptic and global scale have been compared with other estimates based on different methodologies. The influence of the numerical inconsistencies implied by the Lagrangian framework have been reduced significantly by an appropriate exchange model. New insight has been gained in the Lagrangian behaviour of exchanged parcels that relates directly to the chemical forcing induced by stratosphere-troposphere exchange.

There is no commonly accepted definition of the dynamical tropopause (see Chapter 2). The values of potential vorticity generally used vary between 1.5 and 4PVU. Estimates of exchange fluxes are however expected to vary significantly whether the 1.5PVU or the 4PVU surface is used. As noted in Chapter 2, the selection of the best iso-PV surface to mimic a realistic tropopause is not obvious. One thrust of this Chapter is to investigate the possible existence of a preferred iso-PV surface which would minimise exchange, defined in terms of a barrier to vertical mixing.

A Lagrangian estimate of STE for a range of PV-levels provides a novel and alternative perspective on the global diabatic circulation in mid-latitudes. A second theme of the Chapter is to investigate the transport in the upper-troposphere lower-stratosphere from the foregoing perspective.

After a brief description of the methodology, section 7.2 provides a discussion of the preferred tropopause level. Section 7.3 is concerned with large scale transport as inferred from the Lagrangian / PV coordinate framework, while in section 7.4 a link is offered with the usual pressure coordinate viewpoint.

7.1 Methodological Point

The methodology described in Chapter 5 is applied here at several distinct PV-levels (1.5, 2, 3, 5 and 8PVU). The three-dimensional trajectory starting grid (80 km x 80 km x 30 hPa) covers the whole northern hemisphere and extends vertically from 590 to 50 hPa (the output resolution is 3°x3°). The minimum residence time for the exchange selection is taken as $\tau_{crit}$.
Figure 7.1: Annual and zonal mean potential vorticity (bold lines) and potential temperature (thin line) distributions for the period May 1995 to April 1996. Potential temperature contours are shown every 5 K between 280 - 380 K and every 10 K between 380 - 480 K.

ECMWF analysis data (6-hourly T213L31) is used again for the parcel advection and tropopause determination. As can be seen from the annual and zonal mean PV distribution shown in Fig. 7.1, the PV = 8PVU iso-surface extends to almost 50 hPa in the tropics, where the quality of ECMWF data is decreased by uncertainties linked to: the neighbourhood of the top level (10 hPa); the coarse vertical resolution (levels at 10, 30, 50, 70,... hPa); and the general lack of observational data in this region. Exchange in this zone must therefore be interpreted with caution.

### 7.2 Sensitivity of Exchange to the Tropopause Level

Examination of the geographical distributions of exchange shows that the main spatial and seasonal patterns identified for the 2PVU tropopause are also evident at 1.5 and even up till 8PVU, although the intensity tends to decrease with height. The patterns include global belts of upward and downward exchange with maxima over the continents in summer and over the oceans in winter (see Chapter 5).

Yearly-averaged inter PV-levels spatial correlations of exchange estimates are given in Table 7.1 (with the corresponding standard deviations). The persistence of the spatial structures of downward fluxes is very high (correlation > 0.9) for the range 1.5 - 3PVU and decreases progressively above. The upward fluxes are more variable with height and the spatial structures found at 2PVU are almost lost at 5PVU (correlation 0.52). Standard deviations show a weak month-to-month variability of the correlations. Correlations between the net exchange distributions are in line with the combination of downward and upward flux distribution correlations.
Table 7.1: A) Correlation coefficients of the Northern Hemispheric distributions of exchange across the 1.5, 3, 5, 8PVU levels, with the 2PVU level. Values correspond to the annual mean correlations of monthly mean distributions. Standard deviations from the corresponding annual mean are given in bracket. B) Mean projection factors between the geographical exchange distributions across the 1.5, 3, 5, 8PVU levels and the 2PVU level distribution. Mean values and standard deviations are calculated in the same way as for A). See text for details.

Another important feature of the variability of exchange with the PV-level concerns the changes of intensity. To infer the global amplitude of exchange fluxes relative to the ones at 2PVU level, monthly spatial flux distributions $F^{(kPVU)}(x)$ at each level $k$ have been projected onto the 2PVU level distribution $F^{(2PVU)}(x)$ in the grid-point space, using the relation:

$$F^{(kPVU)}(x_j) = \frac{\sum_i F^{(kPVU)}(x_i) F^{(2PVU)}(x_i) F^{(2PVU)}(x_j)}{\left(\sum_i \left(F^{(2PVU)}(x_i)\right)^2\right)^{1/2}}$$

(7.1)

This projected distribution $F^{(kPVU)}(x)$ represents the flux distribution corrected to fit the spatial structures of the 2PVU distribution. The factor between this projection and the 2PVU distribution itself gives an overall approximation of the relative amplitudes of exchange distributions. This approximation is exact only if the spatial distributions are proportional, and becomes less meaningful if they deviate significantly. Annual mean projection factors are given in Table 7.1 with the corresponding standard deviations. They have similar characteristics and orders of magnitudes for both flux directions. The overall flux values decrease monotonically with PV: $\sim 15\%$ between 1.5 and 2PVU, $\sim 30\%$ between 2 and 3PVU, $\sim 60\%$ between 2 and 5PVU, and $\sim 70\%$ between 2 and 8PVU. Again, the month-to-month variability is relatively small.

Looking at the details of geographical distributions, it appears that although the general tendency is to decrease monotonically with height, the rate of decrease can vary significantly with the horizontal location, even within the range 1.5 - 3PVU. To illustrate this, consider in Fig. 7.2 the two downward flux maxima occurring in winter at 1.5 and 3PVU west of Spain and over the Black Sea, respectively. Noting that the scales have been adapted following the projection factors of Table 7.1, it is noteworthy that the former reasonably resembles the overall factor, while the latter is strongly enhanced at 3PVU. On the other hand, as expected from the correlation analysis, the patterns at 8PVU are significantly different from
those at low levels. Note also the large zone of minimum downward exchange at 8PVU over the northern Atlantic, despite the strong exchange activity found there at 1.5 or 3PVU.

It follows from these analyses that the gross spatial structures of STE fluxes are persistent at least between 1.5 and 3PVU and that the difference between levels can be reasonably well approximated via an overall factor. Furthermore, these factors show little month-to-month variability. However, intensity changes with height of some local maxima can deviate significantly from the overall factors. This geographical differentiation becomes relevant for the regional chemistry in regions with large exchange fluxes.

7.3 Zonal Mean View of Exchange in PV Coordinate

In general, the vertical component of the global diabatic circulation of mass can be inferred by representing the zonal and seasonal mean mass flux in a vertical-meridional plane for a specified vertical coordinate. The classical diabatic circulation deduced from the TEM framework is usually cast in pressure coordinate. A conceptually different representation is proposed in Fig. 7.3 (downward fluxes), 7.4 (upward fluxes) and 7.5 (net downward fluxes). The plots represent the mean vertical fluxes across surfaces indexed with the PV, used as vertical coordinate. The mean position of these surfaces is shown in Fig. 7.1. The linkage between the two representations will be discussed in the next section. The conservation of potential vorticity for adiabatic motions renders iso-PV surfaces particularly suitable to estimate the zonally averaged diabatically induced Lagrangian transport. Indeed, the cancellation between large upward and downward fluxes resulting from vertically oscillating adiabatic motions that would occur in a classical zonal mean calculation is avoided when PV coordinate is used. This implies improved estimates at elevations where the vertical adiabatic motion is significant. Together with our Lagrangian methodology, this provides an attractive way of quantifying the vertical component of the global scale tracer circulation at tropopause elevations.

Figure 7.3 shows the seasonal mean zonally integrated downward mass flux. The overall structure has a monotonic decrease with height with a maximum near 40°-50°N in all seasons. The seasonal cycle is more pronounced at lower levels but is almost non-existent above 5PVU. The seasonal maximum occurs in winter and the minimum in summer-autumn.

The corresponding upward fluxes are given in Fig. 7.4. Two maxima are present at low levels in all seasons (but only weakly in summer), and they tend to merge at higher levels to form a maximum between 10° and 40°N with a slow northward decrease. The seasonal variation is strongest at low levels and is again almost absent above 5PVU. The maximum at higher latitudes follows a clear seasonal cycle with larger values in winter and smaller in summer. In this latitudinal sector upward and downward fluxes show a similar seasonal cycle which reflects the Rossby wave-breaking activity (see McIntyre and Palmer 1983, Haynes and McIntyre 1987, Randel et al. 1993). The low latitude maximum has a weaker seasonal cycle in particular at levels above 2PVU. This low latitude band of upward exchange is part of the tropical upwelling and is represented here only partially (the decrease south of 20°N is not realistic). As already discussed in Chapter 5, the flux in this band might be underestimated due to the inaccurate representation of subgrid-scale processes in the
Figure 7.2: Geographical distribution of exchange mass fluxes across various iso-PV surfaces (1.5, 3 and 8PVU) for winter (DJF). The left column represents downward exchange and the right column upward exchange. Unit is [kg km$^{-2}$ s$^{-1}$].
ECMWF data particularly in association with deep cumulonimbus convection. However, some pertinent larger scales tropical features are well captured, e.g. the monsoon-related troposphere-to-stratosphere exchange. Two new flux components can be derived from the basic upward and downward exchange fluxes and enable new insight to be gained: the first is the usual net exchange flux and reveals the large-scale net circulation; the second component is the common part of the upward and downward exchange fluxes which cancels in the netto. This latter component, called hereafter the two-way exchange flux (see also Chapter 5), is large in regions where both upward and downward exchange activities are strong, and therefore represents the large-scale mixing.\(^1\) Note that these two new flux components carry together the same information as the basic fluxes.

Figure 7.5 represents the net exchange flux (positive is downward). Upward and downward fluxes combine to form three distinct zones with: (i) a net upward flux south of ~ 30°N; (ii) a net downward flux from 30°N to 60°N; and (iii) a slightly upward net flux north of 60°N. The latitude where the net flux switches from upward to downward, the “turnaround latitude”\(^2\), is a region of large horizontal gradient of net flux. Net upward flux and net downward flux have roughly comparable magnitudes but experience different seasonal variabilities: the belt of net downward flux is marked by a strong seasonal cycle with maximum in winter and minimum in summer, while the belt of net upward flux shows a much weaker seasonal variability with a minimum in winter. Note that it can be misleading to deduce a “meridional” transport, along iso-PV surfaces, from these seasonal representations due to the seasonally mean vertical motion of iso-PV surfaces. The overall shape of the net vertical flux is very consistent with the general picture of vertically elongated zones of rising and sinking motion expected from the downward control principle (see Chapter 2). It is interesting to note moreover the large vertical gradient in the net upward and downward flux zones. This suggests that vertical transport is composed for a part at least of intense shallow exchanges.

The two-way exchange flux component is shown in Fig. 7.6. The overall structure comprises two maxima of similar intensity, a sharp one near 30°N and a broader around 50° - 60°N (except in summer where they are merged in a spread maximum between 40° - 60°N). Low PV-levels are marked by a distinct seasonal cycle, with a maximum in winter and a minimum in summer, in accord with baroclinic wave-breaking activity in the midlatitudes. At higher levels, the seasonal cycle is reduced and even reversed at 8PVU.

Note furthermore that the rough alignment of two-way exchange contours and PV contours illustrates the effect of potential vorticity gradients on mixing (see also (2.27)). Finally, the similar overall amplitudes found in the two-way exchange flux and net exchange flux indicates that the large-scale mixing can be as significant\(^3\) as the large-scale circulation for the chemical and radiative forcing induced by exchange.

\(^1\)The term “two-way exchange” is used here in a large-scale meaning and do not imply any material link between individual upward and downward exchanges.

\(^2\)Cf Rosenlof (1995) for instance.

\(^3\) Depending on the residence time distribution.
Figure 7.3: Meridional view in PV coordinate of seasonally averaged zonally integrated \textit{downward} mass flux. The unit is in $10^6 \text{[kg s}^{-1} \text{km}^{-1}]$. The vertical axis denotes the PV-level (1.5, 2, 3, 5, 8PVU), and the horizontal axis the latitude. Panel (a) is for winter DJF, (b) for spring MAM, (c) for summer JJA and (d) for autumn SON.
Figure 7.4: Same as Fig. 7.3 but for upward flux.
Figure 7.5: Same as Fig. 7.3 but for net flux. Solid contour lines indicate positive (downward) flux, and dashed lines indicate negative (upward) flux.
Figure 7.6: Same as Fig. 7.3 but for two-way exchange flux.
7.4 Pressure Coordinate View

Analysis of the vertical component of the global mass circulation in the last section has benefited from the adoption of the Lagrangian / PV coordinate framework, and led to the identification of several notable large-scale features.

These estimates of the vertical mass circulation are yielded precisely in the region where usual methods of estimating the diabatic circulation have too large uncertainties\(^4\) (the upper-troposphere lower-stratosphere). Thus, it would be of great interest to use the estimates presented here in a "downward continuation" of existing estimates.

To allow such inferences, the mean exchange fluxes across the range of PV-levels (see for instance Fig. 7.7 (a)) have been interpolated on the corresponding mean iso-PV surfaces in pressure coordinate (correspondence in Fig. 7.7 (b)).

As pointed out by Juckes (1999), the differential vertical motion of iso-PV surfaces and air in the upper-troposphere lower-stratosphere might be such that, especially in the high latitudes, a material increase of the PV value of an air parcel is likely to be typically associated with a descent in pressure coordinate. This means that estimates using a PV coordinate might reveal slightly different patterns on the global scale than those expected from estimates using a pressure coordinate (see also Juckes (2001)).

7.4.1 Net Exchange Flux

Figure 7.7 (a) shows the annual mean net exchange flux calculated and represented in PV coordinate, and Fig. 7.7 (b) the corresponding representation in pressure coordinate. The annual mean eliminates exchange fluxes related to the seasonal variability of the iso-PV surfaces.

Global comparison with results from Elusztkiewicz et al. (1997) (their Fig. 5; see Fig. 2.1) and with results at 100 hPa from Rosenlof (1995) (in particular their Fig. 3) shows that the subtropical upwelling zone and the turnaround latitude are nicely consistent. The turnaround latitude even agrees on the seasonal scale with that of Rosenlof (1995) (their Fig. 12), if the 8PVU iso-surface is taken to represent roughly the 100 hPa level at this latitude. The zone of net downward flux however is much more concentrated to the region south of 70°N in our estimates. The particularly large vertical distance between the 100 hPa and 8PVU iso-surfaces at high latitudes could allow for this difference. This would imply that the net downward motion present at high latitudes near 100 hPa is redirected within the lowermost stratosphere towards the midlatitudes inward the region of net downward flux found in our estimates. On the other hand, the differential motion of the iso-PV surfaces and the air might be able to explain this discrepancy alone. In effect, Juckes (2001) shows estimates of zonally averaged streamfunctions following constant pressure, theta and PV. His vertical mass flux in PV coordinate exhibits a similar general structure to that of Fig. 7.7 (b), with an ascending motion in the high latitudes.

\(^4\)As noted in Chapter 2, the vertical component of the diabatic circulation, being usually estimated relative to constant pressure levels, is subject to uncertainties related to the large variability in the lowermost stratosphere, such estimates are limited to levels well above the tropopause (100 hPa and above).
Figure 7.7: Yearly averaged zonal mean net downward flux, (a) in PV coordinate, and (b) interpolated onto corresponding mean iso-PV surfaces. Solid contour lines indicate positive (downward) fluxes, and dashed lines indicate negative (upward) fluxes. The unit is in $10^6$ [kg s$^{-1}$ km$^{-1}$]. Note that fluxes below 1.5PVU and above 8PVU have not been calculated.

The orientation of iso-PV surfaces in panel (b) of Fig. 7.7 in comparison to isentropes illustrates the discrepancies in the meridional “horizontal” net transports as seen from the iso-PV or the isentropic points of view. For instance, consider the interface between net upward and downward fluxes; this region has a zero cross-PV flux. But on the other hand, the strong upward and downward fluxes produce through mass conservation in a PV-layer a large meridional flux (along iso-PV surfaces) which corresponds to a large cross-isentropic flux. It would therefore be of fundamental interest to assess additionally cross-isentropic exchange with the method proposed here applied to isentropes instead of iso-PV surfaces.

### 7.4.2 Two-way Exchange Flux

Figure 7.8 shows the two-way exchange fluxes represented in pressure coordinate on a seasonal basis. The two flux maxima found in PV coordinate representations (see last section and Fig. 7.6) actually occur in the midlatitude zone of high baroclinic wave activity and in the steepest region of the tropopause close to the subtropical jet stream. The contours of two-way flux in the latter zone have steeper slopes than iso-PV surfaces and the minimum found in PV coordinate between the two maxima is now the natural base of these slopes. It is interesting to note that the maximum of two-way flux in this region occurs in winter, where the slope of iso-PV surfaces is the strongest. On the other hand, two-way flux in the midlatitudes is maximum in a wide meridional belt till 70°N and has significant values reaching the pole. This mixing is clearly related to the baroclinic wave breaking which occurs in the region.
Figure 7.8: Same as Fig. 7.7 (b), but for two-way exchange fluxes. Panel (a) is for winter DJF, (b) for spring MAM, (c) for summer JJA and (d) for autumn SON.
It is interesting to note furthermore that although two-way fluxes contours are reasonably aligned with iso-PV surfaces at low PV-levels, these contours become rather vertically aligned above 5PVU. This suggests that permeability of iso-PV surfaces has two regimes; the first (below 5PVU) is dominantly related to the potential vorticity, while the second (above 5PVU), being not any more related to PV, might possibly be related to the angular momentum. This behaviour seems to reduce the idea of a mixing barrier (where mixing is minimum) to the presence of an enhanced gradient in the mixing activity, without minimum.

7.5 Summary

The analysis has led to suggest a choice of the dynamical tropopause in the range 1.5 to 3 PVU, and furthermore has shown large variations in the exchange intensity in this PV-range, implying the need for a convention.

The PV coordinate perspective has enlightened striking features of the global mass circulation in the upper-troposphere lowermost-stratosphere. Availability of separate upward and downward fluxes has allowed the analysis of two derived fluxes, namely the net and the two-way exchange fluxes. Net exchange fluxes have revealed the two zones of net upward and net downward fluxes of the diabatic circulation as well as their different seasonal variabilities. Symmetry between up and downward flux regions has conferred on the PV coordinate framework a natural place in the analysis. Two-way exchange fluxes have revealed an interesting overall picture composed of two different regimes; below 5PVU, two-way fluxes contours are aligned with potential vorticity, while above they tend to become vertical. Two-way exchange fluxes have amplitudes similar to those found in the net upward and downward flux zones; this enlights the importance of mixing, beside the diabatic circulation, for the global chemistry.
Chapter 8

A One-Year Climatology of Cross-Tropopause O\textsubscript{3} Flux in the Atlantic - European Region

In the previous Chapters various estimates have been derived of cross-tropopause mass fluxes. Stratosphere-to-troposphere exchange of mass implies in general the injection of ozone-rich air into the troposphere which can result in important radiative and chemical effects in the troposphere. The issue of ozone exchange and the subsequent tropospheric forcing has been discussed in several studies (see Introduction), and the stratospheric contribution to the total ozone in the troposphere has been estimated to about 40\% (Roelofs and Lelieveld 1997). The cross-tropopause fluxes of ozone is therefore a key factor for the chemistry and radiative processes in the troposphere. Likewise the upward exchange of mass has an influence upon the stratosphere via injection of tropospheric species, although the injection of ozone into the stratosphere does not engender a chemical forcing due to the high existing background values. However the low ozone concentrations of the injected mass and the subsequent ozone dilution in the stratosphere induce a negative radiative forcing. Upward exchange of ozone is therefore also of interest.

Previous studies of ozone STE have followed three paths. First, observational synoptic-scale studies have concentrated on episodic events, such as tropopause folds, and the ozone exchange has been estimated with the aid of in-situ measurements (aircraft, radiosonde and lidar) (Danielsen 1968, Aancellet et al. 1991, Vaughan et al. 1994, Langford et al. 1996, Langford and Reid 1998). These observational studies often used ozone as a stratospheric tracer to diagnose stratosphere-to-troposphere exchange mass. Secondly, synoptic-scale estimates have also been provided combining a mesoscale numerical model with simplified evaluations of ozone (Ebel et al. 1991, Lamarque and Hess 1994). Finally, general circulation model studies have been conducted using GCMs coupled to chemical models to infer global geographical distributions of STE and their forcing on the troposphere (Roelofs and Lelieveld 1997, Lamarque et al. 1999).

Here an attempt is made to provide detailed estimates of cross-tropopause fluxes of ozone, using our Lagrangian STE methodology and observational in-situ ozone measurements from commercial aircrafts. The Chapter is composed of a brief methodological outline and a
presentation and discussion of the results.

8.1 Methodological Point

Ozone cross-tropopause flux estimates are calculated by combining our mass flux estimates across the 2PVU surface (Chapter 5) with in-situ ozone measurements near the tropopause from the commercial aircrafts involved in the projects NOXAR (Brunner 1998) and MOZAIC (Marenco 1996). Measurements from commercial aircrafts provide one of the few sources for ozone at the tropopause levels that provide a substantial spatial and temporal coverage and a reasonable accuracy.

In a first step, the mean ozone concentration associated with cross-tropopause exchange is derived from the measurements with the following scheme - separately for stratosphere-to-troposphere and troposphere-to-stratosphere exchange: Forward and backward trajectories are computed from all aircraft measurement points from May 1995 to April 1996 with a time-span of 96h and are subjected to our exchange criterion. For the exchange trajectories, the measured ozone values are attributed to the exchange location by assuming the conservation of the ozone volume mixing ratio along the trajectories. The conservation of ozone mixing ratio along the trajectory, between the measurement and the exchange, is reasonable for the chemical lifetime of ozone which is of the order of a month in the upper troposphere - lower stratosphere (Brunner 1998). Then, these ozone mixing ratio are averaged seasonally and spatially on a $3^\circ \times 3^\circ$ grid.

Figure 8.1 represents the geographical distribution of ozone measurement points from the NOXAR and MOZAIC projects. The measurements are concentrated along aircraft routes over Europe and the North Atlantic. The advection of measured ozone concentrations along trajectories directed towards the tropopause permits an enlargement of the geographical coverage of the ozone distributions relatively to the measurement points.

In a second step, these mean ozone concentrations for stratosphere-to-troposphere / troposphere-to-stratosphere exchange are used to specify the ozone content of the cross-tropopause mass fluxes (Chapter 5), which finally provide estimates of ozone fluxes across the tropopause. The value of $\tau_{crit} = 48h$ is used for the critical residence time, according to Chapter 5.

8.2 Results and Discussion

The methodology provides estimates of cross-tropopause ozone fluxes with synoptic-scale details and with a relatively large coverage in the mid-latitudes that extends from the North Atlantic to eastern Europe (see for instance Fig. 8.3). Hereafter, spatially averaged results are discussed to shed light on the seasonal variability, then the geographical variability is examined and finally a comparison with existing estimates from the literature provided.

1 Note that the volume mixing ratio accounts for hydrostatic displacements.
2 Note that only those measurements are considered where the trajectories cross the tropopause within four days prior and after the measurement.
8.2. RESULTS AND DISCUSSION

Figure 8.1: Geographical distribution of in-situ measurements of O\textsubscript{3} from the aircrafts involved in the NOXAR and MOZAIC projects for the period May 1995 to April 1996 (in thousands of measurement points). The scale from 0.5 to 5 thousands of measurements is logarithmic.

<table>
<thead>
<tr>
<th>Season</th>
<th>S→T O\textsubscript{3} flux</th>
<th>O\textsubscript{3} vmr</th>
<th>Mass flux</th>
<th>T→S O\textsubscript{3} flux</th>
<th>O\textsubscript{3} vmr</th>
<th>Mass flux</th>
<th>NET O\textsubscript{3} flux</th>
<th>Mass flux</th>
</tr>
</thead>
<tbody>
<tr>
<td>DJF</td>
<td>42.0</td>
<td>84.8</td>
<td>290</td>
<td>24.7</td>
<td>70.1</td>
<td>211</td>
<td>17</td>
<td>79</td>
</tr>
<tr>
<td>MAM</td>
<td>45.5</td>
<td>118</td>
<td>230</td>
<td>26.3</td>
<td>92.8</td>
<td>169</td>
<td>19</td>
<td>61</td>
</tr>
<tr>
<td>JJA</td>
<td>44.3</td>
<td>125</td>
<td>214</td>
<td>32.3</td>
<td>103</td>
<td>189</td>
<td>12</td>
<td>25</td>
</tr>
<tr>
<td>SON</td>
<td>32.7</td>
<td>83.6</td>
<td>237</td>
<td>22.5</td>
<td>67.8</td>
<td>194</td>
<td>10</td>
<td>43</td>
</tr>
<tr>
<td>Annual</td>
<td>41.1</td>
<td>103</td>
<td>243</td>
<td>26.5</td>
<td>83.4</td>
<td>191</td>
<td>15</td>
<td>52</td>
</tr>
</tbody>
</table>

Table 8.1: Seasonal variability of the mean cross-tropopause O\textsubscript{3} flux ("O\textsubscript{3} flux"), O\textsubscript{3} volume mixing ratio ("O\textsubscript{3} vmr") and the cross-tropopause mass flux ("Mass flux") for stratosphere-to-troposphere (S→T), troposphere-to-stratosphere (T→S), and the NET cross-tropopause flux. The average is proceeded spatially for the domain 60°W - 42°E, 33° - 66°N. O\textsubscript{3} flux is given in 10^{-6} kg s^{-1} km^{-2}, O\textsubscript{3} vmr is in ppbv and Mass flux is in kg s^{-1} km^{-2}.

8.2.1 The Global Seasonal Variability of Cross-Tropopause O\textsubscript{3} Flux

Table 8.1 provides on a spatially averaged basis the ozone fluxes across the tropopause along with the ozone concentration of exchange and the cross-tropopause mass flux. To shed light on the relative weights of these two components throughout the seasons, these figures have been normalised with their annual mean (see Fig. 8.2). Black bars in Fig. 8.2 show the ozone flux across the tropopause, while dark and light grey bars represent respectively the ozone concentration of exchange and the mass flux.

The stratosphere-to-troposphere ozone flux has similar high values during winter, spring and summer, with a small maximum in spring, and lower values in autumn. On the other hand the mass flux, as expected from Chapter 5, has its marked maximum in winter. This difference in the seasonal variabilities of ozone and mass fluxes is explained by the seasonal variability of the ozone concentration of exchange which undergoes a marked maximum in (spring -) summer. The seasonal variability of the cross-tropopause ozone flux is therefore strongly influenced by the seasonal variability of ozone concentration in the lowermost
The troposphere-to-stratosphere ozone flux is also strongly influenced by the ozone concentration of exchange. Ozone and mass flux experience their maximum in summer and winter, respectively. The ozone concentration of troposphere-to-stratosphere exchange, which has a seasonal variability similar to that of the downward exchange, is responsible for this difference. Another interesting aspect is associated with the troposphere-to-stratosphere ozone flux. The stratospheric radiative budget is likely to be forced by the massive injection of air with low ozone values. It can be inferred from Fig. 8.2 that in autumn and winter large mass fluxes with low ozone content will induce a maximum of “negative” radiative effect in the stratosphere.

The net flux of ozone is important to assess the global circulation of ozone and its seasonal variability. The net ozone flux variability is shown on the right panel of Fig. 8.2 along with the net mass flux. The effect of ozone concentration of exchange is to shift the maximum of net ozone flux from winter to spring and the minimum from summer to autumn, in comparison with to the mass exchange.

### 8.2.2 The Geographical Distribution of Cross-Tropopause O₃ Flux

The seasonal mean geographical distributions of ozone cross-tropopause fluxes are represented in Fig. 8.3 separately for downward and upward direction. The geographical variability is strong (in particular for downward fluxes): maxima often reach values five times as large as the background. The relationship between the geographical structures of ozone fluxes and the storm-tracks / continents is not as marked as has been observed for the mass fluxes (Chapter 5), due to the presence of additional structures implied by the ozone concentration of exchange. The spring maximum of stratosphere-to-troposphere ozone flux is due to large values over the North-Atlantic, and is the result of coincident maxima in the mass flux and in the ozone concentration. In contrast, the secondary maximum of ozone flux occurring over eastern Europe in winter is produced purely by a maximum in the ozone.
seasonal variability of the mean O$_3$ cross-tropopause flux in 10$^{-6}$ kg s$^{-1}$ km$^{-2}$. (From Roelofs and Lelieveld 1997.)

Radiative effects in the stratosphere related to troposphere-to-stratosphere exchange are, as noted earlier, not indicated by the upward ozone flux, but rather by the flux of air with low ozone content injected into the stratosphere. To obtain an indication of the significance and variability of this negative forcing, the flux of “missing ozone” has been estimated in the following way: a global concentration of ozone in the lowermost stratosphere of 180 ppbv has been assumed and the difference between the ozone concentration of exchange and this background has been used as the missing concentration of ozone, and a missing ozone flux has been derived as the combination of the missing concentration of ozone and the mass flux. The missing ozone flux is reported geographically in Fig. 8.4. The overall structure possesses a strong south-north gradient and a seasonal cycle with a remarkable maximum in winter and autumn. The magnitudes are similar to those found in the downward ozone fluxes (left panels of Fig. 8.3). This gives a clear indication of the importance of such a negative radiative forcing in the stratosphere.

### Table 8.2: Seasonal variability of the mean O$_3$ cross-tropopause flux in 10$^{-6}$ kg s$^{-1}$ km$^{-2}$.

<table>
<thead>
<tr>
<th>Season</th>
<th>S→T O$_3$ flux</th>
<th>T→S O$_3$ flux</th>
</tr>
</thead>
<tbody>
<tr>
<td>DJF</td>
<td>66.0</td>
<td>25.5</td>
</tr>
<tr>
<td>MAM</td>
<td>78.5</td>
<td>41.5</td>
</tr>
<tr>
<td>JJA</td>
<td>65.5</td>
<td>48.5</td>
</tr>
<tr>
<td>SON</td>
<td>50.5</td>
<td>33.5</td>
</tr>
<tr>
<td>Annual</td>
<td>65</td>
<td>37</td>
</tr>
</tbody>
</table>

8.3 Further Remarks

The comparison of our estimates (Table 8.1) with those of previous studies shows in general a reasonable agreement. In one of the most advanced GCM studies of STE, Roelofs and Lelieveld (1997) used the ECHAM-4 model coupled to a tropospheric chemistry model with a T30 spectral resolution (3.75° x 3.75°) to estimate the global ozone flux across the 3.5 PVU tropopause and to assess chemical effects in the troposphere. Their cross-tropopause ozone flux estimates are reproduced in Table 8.2 on a seasonal basis. The seasonal variability of both flux directions are similar to those of our estimates: maximum of downward (upward) ozone flux in spring (summer). These fluxes are larger than our estimates by a factor of about 1.5, but considering the underlying uncertainties the relative good agreement is encouraging.

With their estimates of the ozone flux, Roelofs and Lelieveld (1997) found a global contribution on the tropospheric ozone from the stratosphere of 40%, and a contribution at the surface between 10% in summer and 60% in winter. The cross-tropopause flux of ozone is therefore a very important source of ozone for the troposphere. The confinement of strong ozone inflows to particular regions (as found in our analysis) can further strengthen the local
Figure 8.3: Geographical distributions of O$_3$ exchange fluxes separately for stratosphere-to-troposphere (left panels) and troposphere-to-stratosphere (right panels), in units of $10^{-6}$ kg s$^{-1}$ km$^{-2}$. (a) is for winter (DJF), (b) for spring (MAM), (c) for summer (JJA) and (d) for autumn (SON).
8.3. FURTHER REMARKS

Figure 8.4: Geographical distributions of poor-ozone mass flux into the stratosphere. The flux of "missing ozone" is represented in units of $10^{-6}$ kg s$^{-1}$ km$^{-2}$. (a) is for winter (DJF), (b) for spring (MAM), (c) for summer (JJA) and (d) for autumn (SON). See text for details.

chemical and radiative forcing in the troposphere that was found in these GCM estimates.

The analysis of cross-tropopause ozone fluxes and their two components, the ozone concentration of exchange and the mass flux, has shown that the ozone concentration field has a significant influence both on the seasonal and spatial variabilities. Therefore, parametrisations of cross-tropopause ozone fluxes should include the variability of ozone concentration. Enhanced downward ozone flux was found during winter, spring and summer with a small maximum in spring, while the maximum in upward ozone fluxes occurred in summer.

Furthermore, a missing ozone flux has been derived to show the potential for a "negative" radiative forcing in the stratosphere induced by troposphere-to-stratosphere exchange. The missing ozone flux turned out to be of the same order of magnitude than the stratosphere-to-troposphere ozone flux. The spatio-temporal variability showed a maximum impact north of 60°N during autumn and winter. To assess quantitatively the significance of these stratospheric and tropospheric radiative perturbations and their possible contribution on the global circulation further investigations are required.

Note finally that the magnitudes of the separate upward and downward fluxes of ozone are both important and therefore that a knowledge of only the net flux (Table 8.1) would certainly be misleading for chemical and radiative purposes.
Chapter 9

An Assessment of the Chemical Forcing due to STE

Dynamical and physical aspects of exchange have been discussed in previous Chapters. Exchange has additionally important implications for the chemistry (and radiation\(^1\)) in the atmosphere because it is the material link between the two quasi-isolated atmospheric layers that are the troposphere and the stratosphere. An injection of tropospheric (stratospheric) constituents into the stratosphere (troposphere) brings foreign species which can force significantly the chemistry in the stratosphere (troposphere). Two well-known examples (cf. Chapter 1) are the injection of stratospheric ozone into the troposphere, and the presence of anthropogenic chlorofluorocarbons (CFCs) in the stratosphere.

On the other hand, the discussion in Chapter 6 has shown that in general the parcels which are exchanged across the tropopause can not be considered as exchanged forever. The mass flux which is still exchanged after a time period \(\tau_{\text{crit}}\) has been empirically formulated as

\[
F(\tau_{\text{crit}}; x, y, t) \simeq (F_0(x, y, t) - F_\infty(x, y, t)) \exp \left( -\ln 2 \frac{\tau_{\text{crit}}}{\tau_{1/2}(x, y, t)} \right) + F_\infty(x, y, t). \tag{9.1}
\]

The characteristic residence time \(\tau_{1/2}\) characterises the overall residence time of the exchange mass flux. \(F_\infty\) represents the "irreversible" exchange mass flux, where "irreversible" corresponds to more than a week. \(F_0\) denotes the exponential extrapolation of \(F(\tau_{\text{crit}})\) for \(\tau_{\text{crit}} = 0\). The analysis has evidenced a significant spatio-temporal variability of the residence time distribution's parameters.

The chemical forcing induced by exchange is clearly influenced by the residence time, and therefore a knowledge restricted to the mass flux at a fixed \(\tau_{\text{crit}}\) can be dramatically insufficient, and in particular for large-scale purposes.

An important implication of this concerns the chemistry evaluated from chemical models coupled to large-scale models. GCMs are generally not able to capture the physical and dynamical processes that are involved in the stratosphere-to-troposphere exchange (cf. Chapter 2). However, global estimates of STE resulting from GCM studies have been extensively

\(^1\)In this study, the focus is placed on the chemical forcing aspects, but the results can easily be extended to radiative purposes.
Figure 9.1: Schematic of the simplified model for the formulation of the forcing on the stratosphere induced by a troposphere-to-stratosphere exchange mass flux. The rate of production of the species C is used as indicator for chemical forcing due to the exchange of the species A, the species B being assumed to be abundant in the stratosphere.

checked and results are encouraging (Mote et al. 1994, Cox et al. 1997, Rind et al. 1999). The correct representation of the residence time distribution which is associated with the STE flux is likely to be more difficult.

The goal of this study is to assess the importance of the residence time distribution on the chemical forcing in the context of our one-year climatology (cf. Chapter 5). In the first part of this Chapter, the indicator for the chemical forcing due to STE is formulated and discussed. The second part of the Chapter concerns the application of the indicator to our one-year climatology (cf. Chapters 5 and 6), and the spatio-temporal variability of the chemical forcing indicator is presented with regard to the time-scale of the chemical lifetime.

9.1 A Simple Indicator for the Chemical Forcing due to STE

9.1.1 Formulation of the Indicator

The aim for an indicator of the chemical forcing due to STE is to provide the potential forcing of the exchange mass flux on the chemistry. Here, this potential forcing is viewed as the rate of production of a new species which is produced by the chemical reaction of the exchanged species with a surrounding constituent.

The simplified model used for the formulation is illustrated in Fig. 9.1 and is derived in Appendix D. The tropospheric species A is injected into the stratosphere with the rate given by the troposphere-to-stratosphere mass flux of A, $F_A$, and the stratospheric reactant species B is assumed abundant and of constant concentration. The first-order two-component chemical reaction of A and B produces the species C:

$$ A + B \rightarrow C $$

(9.2)
The exchange mass flux of \( A \) can be approximated by (9.1)

\[
F_A(\tau_{\text{crit}}) \approx (F_{A0} - F_{A\infty}) \exp\left(-\ln 2 \frac{\tau_{\text{crit}}}{\tau_{A1/2}}\right) + F_{A\infty} \tag{9.3}
\]

\[
\approx F_{A0} (P_{A\infty} + (1 - P_{A\infty}) \exp\left(-\ln 2 \frac{\tau_{\text{crit}}}{\tau_{A1/2}}\right)), \tag{9.4}
\]

where \( F_{A0} \) is the exchange mass flux of \( A \) corresponding to \( \tau_{\text{crit}} = 0 \); \( P_{A\infty} \) is the probability that an exchanged mass element of \( A \) is “irreversibly exchanged”; and \( \tau_{A1/2} \) is the characteristic residence time of the exchange mass flux of \( A \). Note that here the extrapolation of \( F_A(\tau_{\text{crit}}) \) for values of \( \tau_{\text{crit}} \) down to zero is provided by the exponential (9.4), but the absence of “observations” for such values of \( \tau_{\text{crit}} \) implies a possible uncertainty of the method. For simplicity, let’s assume that the flux of \( A \) \( F_A \) can be approached with values for \( P_{A\infty} \) and \( \tau_{A1/2} \) similar to that of the flux of mass \( F \)

\[
F(\tau_{\text{crit}}) \approx F_0 \left( P_{\infty} + (1 - P_{\infty}) \exp\left(-\ln 2 \frac{\tau_{\text{crit}}}{\tau_{1/2}}\right)\right), \tag{9.5}
\]

\[
P_{A\infty} \approx P_{\infty} \tag{9.6}
\]

\[
\tau_{A1/2} \approx \tau_{1/2}. \tag{9.7}
\]

Then, the production of mass of \( C \), \( M_C \), issued from the competition between the residence time of \( A \) and the chemical reaction (9.2) within the stratosphere, can be expressed without the transient terms as:

\[
M_c(t) = F_{A0} \mathcal{E} \, dx \, dy \, t \tag{9.8}
\]

\[
\mathcal{E} = P_{\infty} \left( P_{\infty} + \frac{1 - P_{\infty}}{1 + \frac{\tau_{\text{chem}}}{\tau_{1/2}}} \right), \tag{9.9}
\]

where, \( \tau_{\text{chem}} \) is the chemical lifetime of \( A \) relative to the chemical reaction (9.2).

The quantity \( \mathcal{E} \) can be viewed as the probability that an exchanged mass element finally produces a mass element of \( C \). Therefore, \( \mathcal{E} \) provides a measure of the efficiency of the exchange flux \( F_A \) for the chemical forcing, and is called hereafter forcing efficiency of exchange. This efficiency \( \mathcal{E} \) can be evaluated empirically via \( P_{\infty} \) and \( \tau_{1/2} \) by fitting the dependence of the exchange mass flux \( F \) with an exponential function (cf. Chapter 6). The quantity \( F_{A0} \mathcal{E} \) represents the actual forcing due to the exchange flux of \( A \). The empirical evaluation of \( F_{A0} \mathcal{E} \) requires the knowledge of \( F_{A0} \). Hereafter, the concentration of \( A \) is assumed constant and therefore, the quantity \( F_0 \mathcal{E} \) is evaluated instead of \( F_{A0} \mathcal{E} \).² Note finally that the forcing due to exchange and its efficiency must be evaluated separately for both stratosphere-to-troposphere and troposphere-to-stratosphere exchange fluxes.

### 9.1.2 Discussion of the Forcing Efficiency of Exchange \( \mathcal{E} \)

The forcing efficiency of exchange \( \mathcal{E} \) (9.9) is represented in Fig. 9.2 as a function of its two independent variables, namely \( P_{\infty} \) and the ratio \( \frac{\tau_{\text{chem}}^{\text{chem}}}{\tau_{1/2}} \). Table 9.1 provides the range of

---

²It would be interesting in a further step to evaluate the forcing due to exchange of a specific species like the ozone.
Figure 9.2: Forcing efficiency of exchange $\mathcal{E}$ (z-axis) as a function of $P_\infty$ and $\tau_{1/2}^{\mathrm{chem}}/\tau_{1/2}$. 

Table 9.1: Hemispheric and annual average (with standard deviation in brackets) of the zonal mean values of the parameters $P_\infty$ and $\tau_{1/2}$ relative to the exponential fit functions of the residence time dependence of the exchange mass flux $F(\tau_{\text{crit}})$. $\text{S}\rightarrow\text{T}$ ($\text{T}\rightarrow\text{S}$) denotes stratosphere-to-troposphere (troposphere-to-stratosphere) exchange.

<table>
<thead>
<tr>
<th></th>
<th>$\text{S}\rightarrow\text{T}$</th>
<th>$\text{T}\rightarrow\text{S}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_\infty$</td>
<td>0.076 (0.051)</td>
<td>0.056 (0.025)</td>
</tr>
<tr>
<td>$\tau_{1/2}$ [h]</td>
<td>16.0 (3.4)</td>
<td>16.3 (2.5)</td>
</tr>
</tbody>
</table>

The estimated zonal mean values of $P_\infty$ and $\tau_{1/2}^{\mathrm{chem}}/\tau_{1/2}$ for the northern hemisphere and the time period May 1995 to April 1996. The region of the surface represented in Fig. 9.2 which is relevant here is therefore confined to the values $P_\infty < 0.2$. And the region $0 < \tau_{1/2}^{\mathrm{chem}}/\tau_{1/2} < 1$ corresponds to a chemical lifetime $\tau_{1/2}^{\mathrm{chem}}$ much smaller than $\sim 16$ h, and $\tau_{1/2}^{\mathrm{chem}}/\tau_{1/2} \gg 1$ to $\tau_{1/2}^{\mathrm{chem}} \gg 16$ h. 

The linear (quadratic) dependence of $\mathcal{E}$ on $P_\infty$ for small (large) values of $\tau_{1/2}^{\mathrm{chem}}/\tau_{1/2}$:

$$\mathcal{E}\left(P_\infty, \frac{\tau_{1/2}^{\mathrm{chem}}}{\tau_{1/2}} = 0\right) = P_\infty,$$

$$\mathcal{E}\left(P_\infty, \frac{\tau_{1/2}^{\mathrm{chem}}}{\tau_{1/2}} \rightarrow \infty\right) = P_\infty^2,$$

that can be seen in Fig. 9.2, implicates a reduced sensitivity of $\mathcal{E}$ with $P_\infty$ for large values of $\tau_{1/2}^{\mathrm{chem}}/\tau_{1/2}$ and small values of $P_\infty$. 

The dependence of $\mathcal{E}$ on $\tau_{1/2}^{\mathrm{chem}}/\tau_{1/2}$ for small and large chemical lifetimes can be approx-
9.1. A SIMPLE INDICATOR FOR THE FORCING

Figure 9.3: Forcing efficiency of exchange $E$ as a function of $\tau_{1/2}^{chem}/\tau_{1/2}$ for three values of $P_\infty$: 0.05, 0.5, 0.95. Dotted lines: approximations (9.13) and (9.15). Dashed lines: values for $\tau_{1/2}^{chem}/\tau_{1/2} = 0$ and $\to \infty$. See text for details.

imated by the first-order Taylor series of $E$:

\[
\frac{\tau_{1/2}^{chem}}{\tau_{1/2}} \ll 1
\]

\[
\Rightarrow E \simeq P_\infty + (1 - P_\infty) \left( 1 - \frac{\tau_{1/2}^{chem}}{\tau_{1/2}} + \mathcal{O}\left(\frac{\tau_{1/2}^{chem}}{\tau_{1/2}}\right)^2 \right), \tag{9.13}
\]

\[
\frac{\tau_{1/2}^{chem}}{\tau_{1/2}} \gg 1 \iff \frac{\tau_{1/2}}{\tau_{1/2}^{chem}} \ll 1
\]

\[
\Rightarrow E \simeq P_\infty + (1 - P_\infty) \left( \frac{\tau_{1/2}^{chem}}{\tau_{1/2}} + \mathcal{O}\left(\frac{\tau_{1/2}}{\tau_{1/2}^{chem}}\right)^2 \right) \tag{9.14}
\]

Hence, for a fixed $\tau_{1/2}^{chem}$, the dependence of $E$ on $\tau_{1/2}$ is $\sim 1/\tau_{1/2}$ for $\tau_{1/2}^{chem} \ll \tau_{1/2}$, but is $\sim \tau_{1/2}$ for $\tau_{1/2}^{chem} \gg \tau_{1/2}$. These dependences are illustrated in Fig. 9.3 for three different values of $P_\infty$.

For small values of $P_\infty$ in the range of those estimated in our one-year climatology (cf. Table 9.1), the expression of $E$ can be further simplified by removing second and higher order terms:

\[
\frac{\tau_{1/2}^{chem}}{\tau_{1/2}} \ll 1, \ P_\infty \ll 1 \Rightarrow E \simeq P_\infty \left[ 1 - \frac{\tau_{1/2}^{chem}}{\tau_{1/2}} + \mathcal{O}^2 \right] \tag{9.16}
\]

\[
\frac{\tau_{1/2}^{chem}}{\tau_{1/2}} \gg 1, \ P_\infty \ll 1 \Rightarrow E \simeq P_\infty \left[ P_\infty + \frac{\tau_{1/2}^{chem}}{\tau_{1/2}} + \mathcal{O}^2 \right]. \tag{9.17}
\]
This shows that the forcing efficiency of exchange is sensitive to the residence time distribution for any chemical lifetime. For long-living chemical species, the forcing due to exchange is the combination of the irreversible exchange flux component $P_\infty$ with the ratio $\tau_{1/2}/\tau_{chem}^{1/2}$. For short-living chemical species, the ratio $\tau_{chem}/\tau_{1/2}$ is dominated by the constant value 1, and hence, $E$ does not depend on $\tau_{1/2}$ anymore (exchanged parcels reside enough time to experience almost certainly a chemical reaction). These dependences shed light on the possible effects of a spatio-temporal variability of the residence time distribution on the global chemistry.

9.2 Application to the One-Year Climatology

The evaluation of the forcing efficiency of exchange $E$ in the context of our one-year climatology is based on the prior evaluation of $P_\infty$ and $\tau_{1/2}$ via the expression (9.9). The estimation of $P_\infty$ and $\tau_{1/2}$ follows from the calculation of the exponential fits of the dependence on $\tau_{crit}$ of $F(\tau_{crit}; x, y, t)$ for every grid-square ($3^\circ \times 3^\circ$) of monthly mean geographical distributions of exchange estimates, as discussed in Chapter 6.

Zonally averaged estimates of $P_\infty$ and $\tau_{1/2}$ are provided in Fig. 9.4 for the two directions of exchange. $P_\infty$ (Fig. 9.4 a) and $\tau_{1/2}$ (Fig. 9.4 b) possess similar spatio-temporal structures for both directions of exchange: a marked seasonal cycle with the minimum in late autumn - early winter; and the meridional profile has a maximum in the midlatitudes for stratosphere-to-troposphere (S→T) exchange, and a general northward gradient for troposphere-to-stratosphere (T→S) exchange. The main difference between $P_\infty$ and $\tau_{1/2}$ is the meridional variability which is much stronger for $P_\infty$.

9.2.1 Estimates of $E$ for Long-Living Species ($\tau_{chem}^{1/2} \gg \tau_{1/2}$)

The zonally averaged forcing efficiency of exchange $E$ estimated with (9.9) and $\tau_{chem}^{1/2} = 1$ week, is provided in Fig. 9.5 (a) for the two directions of exchange. In the S→T case, a marked midlatitude belt of relative large values of $E$, extended to the north pole region in summer, contrasts with the very small values south to $\sim 35^\circ$N. For T→S exchange, the meridional profile is smoother and very low values of $E$ occur in the subtropics during the period late autumn - early winter.

The overall spatio-temporal patterns are similar to that of $P_\infty$ (Fig. 9.4). The relationship between $E$ and $P_\infty$ is evidenced by the approximative expression (9.17) of $E$ for long-living species and small $P_\infty$. The relative weights of the two terms $P_\infty$ and $P_\infty\tau_{1/2}$ are determined by the value of $\tau_{chem}^{1/2}$ - a value $\tau_{chem}^{1/2} = 150$ h ($210$ h) would equalise the weights for the S→T (T→S) case.

9.2.2 Estimates of $E$ for Short-Living Species ($\tau_{chem}^{1/2} \ll \tau_{1/2}$)

Fig. 9.5 (b) shows the forcing efficiency of exchange estimated with (9.9) and $\tau_{chem}^{1/2} = 1$ hour, for the two directions of exchange. Again, the similarity of these patterns with that of $P_\infty$
9.2. APPLICATION TO THE ONE-YEAR CLIMATOLOGY

Figure 9.4: Zonally averaged parameters, (a) $P_\infty$ [a.u.], and (b) $\tau_{1/2}$ [h], of the exponential fit of the dependence on $r_{\text{crit}}$ of the exchange mass flux $F(\tau_{\text{crit}}; x, y, t)$ (cf. (9.1)). X-axis is the month, starting in May 1995 and ending in April 1996; and y-axis is the latitude. $S \rightarrow T$ ($T \rightarrow S$) denotes stratosphere-to-troposphere (troposphere-to-stratosphere) exchange. Dash-dotted lines represent the statistical significance of the exponential fits (contours at 0.9 and 0.95) (cf. Chapter 6 for details).

is evident from the approximated form of $\mathcal{E}$ of (9.16) - the second term in the bracket being dominated by the constant term.

Compared to the estimates of $\mathcal{E}$ with $\tau_{\text{chem}}^{1/2} = 1$ week, the meridional gradient is smoother, the values in the subtropics are relatively enhanced, and the overall order of magnitude of $\mathcal{E}$ is about five times as large.

9.2.3 Estimates of $F_0 \mathcal{E}$

The estimates of $F_0$ necessary for the evaluation of the forcing due to exchange $F_0 \mathcal{E}$ is provided in Fig. 9.6. This quantity corresponds to the Eulerian exchange mass flux (i.e. $F(\tau_{\text{crit}} = 0, x, y, t)$) as extrapolated from the “observed” values of $\tau_{\text{crit}}$ (24 h - 96 h) to $\tau_{\text{crit}} = 0$, via (9.1). The values of $F_0$ for $S \rightarrow T$ and $T \rightarrow S$ have very similar spatio-temporal patterns and orders of magnitudes. Their annual cycle are apparent only in the subtropical region and have the maximum winter and the minimum in summer. The meridional profiles possess a maximum in the subtropics and show a general decrease towards the north.

Finally, the spatio-temporal representation of the forcing due to exchange $F_0 \mathcal{E}$ is provided
Figure 9.5: Zonally averaged forcing efficiency of exchange $\mathcal{E}$ [a.u.] calculated with (9.9) for (a) $\tau_{1/2}^{\text{chem}} = 1$ week and (b) $\tau_{1/2}^{\text{chem}} = 1$ hour. X-axis is the month, starting in May 1995 and ending in April 1996; and y-axis is the latitude. Dash-dotted lines represent the statistical significance of the exponential fits (contours at 0.9 and 0.95).

Figure 9.6: Zonally integrated exchange mass flux $F_0$ (exponential extrapolation of $F(\tau_{\text{crit}}; x, y, t)$ at $\tau_{\text{crit}} = 0$), in $10^6$ kg s$^{-1}$ per 1km broad zonal band. X-axis is the month, starting in May 1995 and ending in April 1996; and y-axis is the latitude. Dash-dotted lines represent the statistical significance of the exponential fits (contours at 0.9 and 0.95).
Figure 9.7: Zonally integrated forcing due to exchange $F_0 \mathcal{E}$ for (a) $\tau_{1/2}^{chem} = 1$ week and (b) $\tau_{1/2}^{chem} = 1$ hour, in $10^6$ kg s$^{-1}$ per 1km broad zonal band. X-axis is the month, starting in May 1995 and ending in April 1996; and y-axis is the latitude. Dash-dotted lines represent the statistical significance of the exponential fits (contours at 0.9 and 0.95).

in Fig. 9.7 for the two chemical lifetimes $\tau_{1/2}^{chem}$: 1 week (a) and 1 hour (b). The forcing due to S→T exchange has an annual cycle with the maximum during winter - spring and minimum during summer - autumn, and is confined in the midlatitude belt and has very low values in the subtropics in particular for the long-living species. The forcing due to T→S exchange is significant at all latitudes, and has no clear annual cycle apart from a distinct minimum in late autumn - early winter in the subtropics. The sensitivity of the overall patterns to the chemical lifetime $\tau_{1/2}^{chem}$ is generally weak.

For both values of $\tau_{1/2}^{chem}$, the comparison with $F_0$ (Fig. 9.6) and $\mathcal{E}$ (Fig. 9.5) demonstrates that the residence time distribution associated with the exchange mass flux plays a central role for the chemical forcing due to the exchange mass flux.

9.3 Summary

A simple indicator for the chemical forcing due to exchange $F_0 \mathcal{E}$ and its efficiency $\mathcal{E}$ has been formulated by making use of the empirical residence time distribution associated to the exchange mass flux which has been proposed in Chapter 6. The forcing efficiency of exchange $\mathcal{E}$ is determined by the irreversible component $P_\infty$ and the characteristic residence time $\tau_{1/2}$.
of the exchange flux and depends on the chemical lifetime $\tau_{1/2}^{chem}$ of the exchanged species. The chemical forcing due to exchange $F_0 \mathcal{E}$ is assessed by the product of this efficiency with the Eulerian exchange flux $F_0$ as extrapolated for $\tau_{crit} = 0$.

The analysis has demonstrated that the chemical forcing due to exchange is strongly influenced by the residence time distribution via the term $\mathcal{E}$ for all chemical lifetimes. The distinct annual cycle (maximum in spring - summer, minimum in autumn - winter) and meridional profile (midlatitude belt for $S \rightarrow T$, and northward gradient for $T \rightarrow S$) of the latter, which are related to those of $P_\infty$ and $\tau_{1/2}$, strongly affects the annual cycle and meridional profile of the forcing due to exchange $F_0 \mathcal{E}$.

The forcing due to $S \rightarrow T$ exchange has a clear seasonal cycle with a maximum in winter - spring and a minimum in summer - autumn. This forcing is strong within the midlatitude belt and very weak in the subtropics - even though $F_0$ has its maximum in the subtropics. The forcing due to $T \rightarrow S$ exchange is more homogeneous annually and meridionally, apart from a distinct minimum in the subtropics in late autumn - early winter.

The sensitivity of the forcing efficiency $\mathcal{E}$ to the chemical lifetime $\tau_{1/2}^{chem}$ is significant on the overall amplitude of $\mathcal{E}$ but is weak on the overall spatio-temporal patterns.

The main caveat of this study is the unavailability of estimates of $F (\tau_{crit}; x, y, t)$ for $\tau_{crit} < 24$ h, and the subsequent use of the exponential extrapolation for these values of $\tau_{crit}$ without "observational" confirmation. Additionally, the representativity of all spatio-temporal patterns found here for other years is subject to the interannual variability. Finally, the simple indicator formulated here has demonstrated the importance of the residence time distribution on the related chemical forcing, but might be too restrictive for accurate evaluations of the chemical forcing.
Chapter 10

Final Discussion and Further Investigations

In this study, the analysis and quantification of stratosphere-troposphere exchange has been performed within the Lagrangian framework. The Lagrangian diagnostic method has been developed with the goal to resolve the small-scale processes relevant to STE and to enable global coverage estimates. The use of the Lagrangian framework was motivated by two evidences. One is the difficulties encountered with Eulerian methods when attempting to quantify STE on a global scale by resolving the small-scale processes relevant to STE. The second is the intrinsic Lagrangian nature of the forcing induced by STE.

Throughout the various analyses presented in this study, the Lagrangian method developed and applied here has proven important potentials for both the analysis and quantification of STE in the extratropics, on both hemispheric and synoptic scales. The results have revealed several striking properties of the cross-tropopause exchange flow.

- Meso-scale analysis of physical processes responsible for exchange has benefited from the three-dimensional representation of exchange trajectories and from the trajectory analysis, and has led to the identification of subtle processes. In particular, it has been shown that the preliminary injection of moist tropospheric air into a cut-off low can lead to an enhanced stratosphere-to-troposphere exchange caused by pure radiative processes, and thereby rapidity the mixing of the cut-off.

- Climatological analysis of the rapid transport (involving STE) between the stratosphere and the boundary layer has revealed a strong zonal asymmetry. Emissions from the near Japan and the American east coast enter much more frequently the stratosphere than emissions from other areas including Europe. And injection of stratospheric ozone in the boundary layer is generally larger over oceans than over continents.

- The residence time distribution of the exchange mass flux has been established empirically: it possesses an strongly decreasing exponential shape which varies spatio-temporally. The residence time characteristics represent an intrinsic Lagrangian property of the exchange mass flux. Furthermore, this property has been shown to have determinant implications for the chemical and radiative forcing due to STE.

- Net exchange mass flux has been estimated across a range of iso-PV surfaces in the upper-troposphere and lowermost stratosphere. A monotonic decrease with the PV-level was
found. Two distinct meridional belts of up- and downward net flux were present at all PV-levels and have been interpreted as the signature of the Brewer-Dobson circulation near the tropopause level.

- The symmetric two-way exchange flux has been estimated. The two-way exchange "activity" (averaging zonally) is uniformly large north of 40°N without significant seasonal variability. This flux possesses a monotonic vertical gradient with the PV-level, with low values at 5PVU and above. The amplitude of the two-way flux is comparable to that of the net flux and has therefore significant consequences on the global forcing due to exchange.

- Analysis of correlations between exchange fluxes across several PV-levels has shown that the iso-PV surfaces between 1.5 and 3PVU have similar geographical structures but overall amplitudes vary dramatically. This is relevant for the choice of the dynamical tropopause.

- Ozone mass exchange has been quantified from in-situ airborne measurements. It was shown that the seasonal cycle of ozone flux is substantially shifted by the seasonal variability of the ozone concentration.

The quantifications of stratosphere-troposphere exchange of mass, ozone on the hemispheric, synoptic scale have been compared with independent studies, based on the residual mean circulation, GCM coupled to chemical model, case studies using different methods, and a good agreement has been obtained for the amplitudes and seasonal cycles. These agreements confirm the quantification capability of the method, while the mesoscale and climatological analyses have provided confidence in the qualitative aspects of the results.

The application of the method to the ECMWF data set implies some limitations which are related to non-resolved processes in the ECMWF model. Of particular relevance in the extratropics might be the deep convection occurring in summer. The other limitation concerns the representativity of the one-year climatology regarding the inter-annual variability. Note that current work at the ETHZ within the EU project STACCATO, aims at applying the method to a 15-years climatology. Numerical errors related to the accuracy / convergence of the trajectory algorithm (see Appendix C) have been argued to be inexistent with the EM model output and weak with the ECMWF data set. This argumentation could be at least qualitatively checked in a further step by comparing estimates of STE using the high resolution model data and directly the ECMWF data set.

Several possible further investigations have been suggested throughout the Chapters which could provide further interesting insights.

- The Eulerian method proposed in Chapter 2 which is based on the new and generalised formulation of the mass flux across a PV contour could be applied to the mesoscale model data set, and the results could be compared to the estimates using the Lagrangian method. Qualitative inferences have already been performed and are very encouraging. Comparisons between a reliable Eulerian method and the Lagrangian method developed here could provide useful information of the exchange mass flux for $\tau_{crit} \approx 0$.

- The use of the HRM model instead of the EM model (Chapter 3) for the case study would permit to quantify the diffusive and diabatic components of the exchange mass flux.

- The application of the method developed here to exchange across isentropic surfaces (using three-dimensional wind fields) would provide complementary information on the large-scale mixing that have been evidenced here across iso-PV surfaces (Chapter 7).
Appendix A

Generalised Cross-Tropopause Flux Formulation

A.1 Preliminaries

The governing equations of a fluid in a rotating environment can be expressed as

\[
\frac{Du}{Dt} + 2\Omega \wedge u = -\frac{1}{\rho} \nabla p - \nabla \Phi + F \quad (A.1)
\]

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0 \quad (A.2)
\]

\[
\frac{D\theta}{Dt} = \dot{\theta} \quad (A.3)
\]

\(u\) is the flow velocity, \(\rho\) the mass density, \(\Omega\) is the angular velocity of the earth, \(\Phi\) represents the geopotential \(g_z\) (or the potential of any conservative body forces) and the non-conservative terms \(F\) and \(\theta\) are the friction and the diabatic heating sources.

The material rate of potential vorticity (PV) \(Q = 1/\rho \omega \cdot \nabla \theta\), where \(\omega = 2\Omega + \nabla \wedge u\) is generally given in the form

\[
\frac{DQ}{Dt} = \frac{1}{\rho} \{\omega \cdot \nabla \dot{\theta} + \nabla \wedge F \cdot \nabla \theta\} \quad (A.4)
\]

Following Haynes and McIntyre (1987; 1990), a "potential vorticity substance" (PVS) can be introduced as \(\rho Q\), and a conservative form of the latter equation can be written in the general case

\[
\frac{\partial \rho Q}{\partial t} + \nabla \cdot \mathbf{J}_{PVS} = 0 \quad (A.5)
\]

\[
\mathbf{J}_{PVS} = \mathbf{J}_{adv} + \mathbf{J}_{na} \quad (A.6)
\]

\[
\mathbf{J}_{adv} = \rho Q u \quad (A.7)
\]

\[
\mathbf{J}_{na} = -\omega \dot{\theta} - F \wedge \nabla \theta \quad (A.8)
\]

\[\dot{\theta} = \rho Q u^{na} \quad (A.9)\]
Schär (1993) explored an other way to express the PVS flux $J^{PVS}$ based on the Bernouilli theorem. The Bernouilli function is defined as

$$B = h + \frac{1}{2}u^2 + \Phi$$  \hspace{1cm} (A.10)

$$\approx c_p T + \frac{1}{2}u^2 + gz$$  \hspace{1cm} (A.11)

and the PVS flux becomes

$$J^{PVS} = \nabla \theta \wedge \left( \nabla B + \frac{\partial u}{\partial t} \right) - \omega \frac{\partial \theta}{\partial t}$$  \hspace{1cm} (A.12)

### A.2 Time Variation of the Mass Enclosed by Boundaries Defined by Fixed (in Time) PV Values

Let $D(t)$ be a volume defined by given PV values at its boundaries, $dV$ an elementary volume and $M(t)$ the mass inside of $D(t)$.

$$M = \int_{D(t)} \rho dV$$  \hspace{1cm} (A.13)

The transport theorem (e.g. Dutton 1976) allows to express the time-derivative of $M$ as

$$\frac{\partial M}{\partial t} = \int_{D(t)} \frac{\partial \rho}{\partial t} dV + \int_{\partial D(t)} \rho w \cdot \eta d\sigma$$  \hspace{1cm} (A.14)

with $\eta =$ vector normal to the surface $\partial D(t)$, pointing outwards, $w =$ velocity vector field of the boundary $\partial D(t)$.

By using the mass continuity equation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0$$  \hspace{1cm} (A.15)

one can write

$$\frac{\partial M}{\partial t} = - \int_{D(t)} \nabla \cdot (\rho u) dV + \int_{\partial D(t)} \rho w \cdot \eta d\sigma$$  \hspace{1cm} (A.16)

$$= - \int_{\partial D(t)} \rho u \cdot \eta d\sigma + \int_{\partial D(t)} \rho w \cdot \eta d\sigma$$  \hspace{1cm} (A.17)

$$= \int_{\partial D(t)} \rho (w - u) \cdot \eta d\sigma$$  \hspace{1cm} (A.18)

Taking the lowest boundary as the tropopause, the flux between the stratosphere and the troposphere is:

$$F = - \int_S \rho (w - u) \cdot \eta d\sigma$$  \hspace{1cm} (A.19)

with the conventions:

$$F \begin{cases} > 0 \Leftrightarrow S \to T, & \eta = - \frac{\nabla Q}{|\nabla Q|} \\ < 0 \Leftrightarrow T \to S \end{cases}$$  \hspace{1cm} (A.20)
A.3 General Expression for $w$

The material derivative of the potential vorticity $Q$ expresses the rate of change of $Q$ following an air parcel

$$\frac{DQ}{Dt} = \frac{\partial Q}{\partial t} + u \cdot \nabla Q = \{\text{sources of } Q\} \quad (A.21)$$

whereas, by definition of $w$, the "material" derivative of $Q$ following a virtual parcel moving with $w$ will be zero

$$\frac{\partial Q}{\partial t} + w \cdot \nabla Q = 0 \quad (A.22)$$

It follows that

$$w = \frac{-\partial Q}{\partial t} \frac{1}{|\nabla Q|^2} \nabla Q + \chi \wedge \nabla Q, \quad \forall \chi \in \mathbb{R}^3 \quad (A.23)$$

A.4 General Expression of the Flux

Making use of the two precedent results concerning $F$ and $w$, as well as the definition for $\eta$ it comes

$$F = -\int_{\sigma} \rho(w - u) \cdot \eta d\sigma \quad (A.24)$$

$$= -\int_{\sigma} \rho \frac{1}{|\nabla Q|} \left( \frac{\partial Q}{\partial t} + u \cdot \nabla Q \right) d\sigma \quad (A.25)$$

$$F = -\int_{\sigma} \rho \frac{1}{|\nabla Q|} \frac{DQ}{Dt} d\sigma \quad (A.26)$$

A.5 Diverse Expressions for the Flux

Using the continuity equation for $\rho Q$ together with the one for $\rho$,

$$\frac{\partial \rho Q}{\partial t} + \nabla \cdot (J^{\text{adv}} + J^{\text{na}}) = 0 \quad (A.27)$$

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0 \quad (A.28)$$

we can write after some manipulations:

$$\frac{DQ}{Dt} = -\frac{1}{\rho} \nabla \cdot J^{\text{na}} \quad (A.29)$$

Thus,

$$F = \int_{\sigma} \frac{\nabla \cdot J^{\text{na}}}{|\nabla Q|} d\sigma \quad (A.30)$$
And
\[ \nabla \cdot J^{na} = -\rho \frac{DQ}{Dt} \]  
  \[ = -\left( \frac{\partial \rho Q}{\partial t} + \nabla \cdot J^{adv} \right) \]  
  \[ = -\nabla \cdot \left( \omega \dot{\theta} + F \wedge \nabla \theta \right) \]  
  \[ = -\nabla \cdot \left( J^{adv} + \omega \frac{\partial \theta}{\partial t} - \nabla \theta \wedge \left( \nabla B + \frac{\partial u}{\partial t} \right) \right) \]  
  \[ = -\left( \omega \cdot \nabla \dot{\theta} + \nabla \wedge F \cdot \nabla \theta \right) \]  

A.6 Another Expression for \( w \)

Rewriting the continuity of the PV-substance
\[ \frac{\partial \rho Q}{\partial t} + \nabla \cdot \left( J^{adv} + J^{na} \right) = 0 \]  

in the form
\[ \frac{\partial Q}{\partial t} + w \cdot \nabla Q = 0 \]

it comes
\[ \frac{\partial Q}{\partial t} + (u + u^{na}) \cdot \nabla Q + \frac{Q}{\rho} \nabla \cdot (\rho u^{na}) = 0 \]  
  \[ \frac{\partial Q}{\partial t} + \left\{ u + u^{na} + \left( \frac{Q}{\rho} \nabla \cdot (\rho u^{na}) \right) \right\} \frac{\nabla Q}{|\nabla Q|^2} + \chi \wedge \nabla Q \cdot \nabla Q = 0 \]  

Thus
\[ w = u + u^{na} + \left( \frac{Q}{\rho} \nabla \cdot (\rho u^{na}) \right) \frac{\nabla Q}{|\nabla Q|^2} + \chi \wedge \nabla Q; \quad \forall \chi \in \mathbb{R}^3 \]
Appendix B

Analyses of the Method

B.1 Starting Grid Analysis

The choice of an appropriate starting grid\(^1\) for the trajectory computation is an important part of our methodology (see section 3.4). The main goal is the optimisation of the grid parameters such that the ultimate estimation quality is acceptable while the computing time requirement is sustainable with the available computation facilities.

B.1.1 Starting Grid Boundaries

A first optimisation procedure concerns the choice of the starting grid boundaries (see Fig. 3.4). The set of trajectories which are launched from the grid-points of the starting grid is supposed to represent the flow in the region of the tropopause in the diagnosis domain continuously during the period \(T\). To determine the boundaries, a reverse approach has been used. Trajectories have been started within a layer of 100hPa around the tropopause inside the diagnosis domain, and calculated backwards for a period \(T\). Then, the box boundaries have been subjectively chosen in order to keep the part of trajectories originating outside the boundaries below 5\% of the total amount of trajectories. Table B.1 gives final results for several values of \(T\). The proportion of lost trajectories is the ratio between those originating outside and those inside, while the proportion of lost volumes represent the ratio between trajectory points outside and inside. This latter error gives, through mass conservation, an estimation of the total effect of lost trajectories for the period \(T\). The threshold of 5\% lost trajectories implicates lost volumes below 2.5\%, and therefore undesired related effects are supposed to be small when using these starting grid boundaries.

B.1.2 Starting Grid Resolution

It is of first importance to have a clear idea of the sensitivity of STE estimates to the starting grid parameters. A first reason is the understanding of the link between resulting estimations and characteristics of the elementary volumes which are represented by trajectories. The

\(^1\)The starting grid refers here as in Chapter 3, to the regular three-dimensional grid whose grid-points coincide with the points where trajectories are started.
second is due to the large increase of the required computer time when the resolution of the starting grid is increased. For example, an increase by a factor 2 of each component of the starting grid leads to a factor $2^3$ on the number of computed trajectories and on the computer time.

The starting grid ($\Delta x^{sg}, \Delta y^{sg}, \Delta p^{sg}, T$) characterises the elementary masses ($dM = 1/g\Delta x^{sg} \Delta y^{sg} \Delta p^{sg}$) which are advected along the trajectories during $T$. Each trajectory crossing the tropopause and satisfying the conditions discussed in section 3.3, represents an exchange of mass $dM$. The resulting exchange mass can be represented on a horizontal geographical map of exchange with a resolution ($\Delta x^{gm}, \Delta y^{gm}, \Delta t^{gm}$). In the whole study it is assumed that $\Delta t^{gm} = T$, so that the geographical map of exchange have the highest available temporal resolution. The elementary area, or the pixel, of the geographical map of exchange is denoted by $\Delta A = \Delta x^{gm} \times \Delta y^{gm}$. Furthermore, horizontal isotropy of the starting grid ($\Delta x^{sg} = \Delta y^{sg}$) is assumed to be justified by the zonal highly asymmetric nature of the synoptic perturbation.

The discussion in section 3.3 led to the definition of significant exchanges as being a further selection from a basic set of preliminary exchange events. In the particular context of this sensitivity study, the exchange is understood as the preliminary exchange$^2$. This assumption permits an important computer-time saving (the trajectory prolongation is not performed), and thereby allows to investigate higher starting grid resolutions. The results of this analysis will be reasonably applicable to the exchange estimates of Chapters 3 and 4.

Table B.2 summaries the ranges for the variations of the parameters $\Delta x^{sg} = \Delta y^{sg}, \Delta p^{sg}, T$. The smallest value are ($T_0=6$ h, $\Delta x_0^{sg} = \Delta y_0^{sg}=0.25^\circ$, $\Delta p_0^{sg}=5$hPa). Hereafter, by convention the zonal component of the starting grid, given in $^\circ$, is normalised such that $1^\circ$ in longitude has the same actual length than $1^\circ$ in latitude.

The following analysis is structured downscale, starting with the overall aspects and ending with detailed geographical structures. But first of all, an outlook which emphasises the main results of the analysis is provided.

$^2$Referring, as defined in section 3.3, to the events defined by a trajectory crossing the tropopause
Outlook

The Lagrangian estimation of geographical distributions of exchange mass and the representation in a pixelised map bring several questions:

(i) what values of $T$ are reasonable for a case study;

(ii) what is the optimised elementary mass $dM$ represented by a trajectory, and what is the optimum ratio between its horizontal and vertical sizes, in order to minimise the computational effort;

(iii) what resolution would be reasonable for the map of estimated exchange.

This analysis provides the following answers:

(i) If a temporal continuity of the exchange maps is desired, $T$ should not overpass 12h.

(ii) Given a resolution for the exchange map $\Delta A$ and $T$, the elementary mass $dM$ must be chosen to respect the condition $R > 10$ in Fig. B.6 ($R$ is introduced below). The horizontal and vertical components of the elementary mass volume should be chosen knowing that estimates are more sensitive to the vertical component (Figs. B.7, B.9 and B.10).

(iii) The relation between the resolution of the exchange map and the elementary volume mass also provides the relation between the resolution of the exchange map and the computational effort. The resolution of maps can therefore be chosen with regard to the necessary computational effort via Fig. B.6.

Overall exchange estimation

The integrated mass exchange for the full available period (96h) and the whole diagnosis domain is represented in Fig. B.1 as a function of $(\Delta x^g = \Delta y^g, \Delta p^g)$, for the smallest and largest $T$ values ($T=6h, 48h$). The exchange amount has been normalised by the mass element $dM_0 = 1/g\Delta x_0^g\Delta y_0^g\Delta p_0^g$, so that the represented values correspond to the number of elementary mass volumes.

Both panels show that in spite of the wide range of $\Delta x^g$ and $\Delta p^g$, the total exchange amount does not vary more than $\sim 10\%$. They also show that an increase of $T$ does only remove a part of the variability without changing the total magnitude. The maps for $T=12$ and $T=24$ (not shown) show intermediate features. As a consequence, $dM$ and $T$ can be varied in a large range without introducing a bias in the resulting estimates.

The variability is dominated by the vertical component, at least for low values of $T$. This dependency is related to the low vertical mobility of parcels. Figure B.2 shows the integrated distributions of horizontal and vertical distances a parcel travels during a period $T = 6, 12, 24, 48h$, computed from this case study. It is shown that half of the trajectories can rise/sink by 5hPa within $T=6h$, and by 20hPa within $T=48h$. Thus, for large values of $\Delta p^g$ relative to the parcel's vertical mobility, a non-negligible part of the vertical layer is not visited by the computed parcels, which are nevertheless supposed to represent the full vertical scale. The consequence is, as seen in Fig. B.1, a growing variability of the estimates as $\Delta p^g$ increases.

The horizontal component of the starting grid is likely to lead to much less variability (at least for the range of grids chosen here) because, as can be seen from Fig. B.2, more than 90% of the parcels are able to travel a distance corresponding to $1°$ within $T=6h$, and 99% within $T=48h$. 
Figure B.1: Total number of exchanged elementary mass volumes as a function of \((\Delta x^g = \Delta y^g, \Delta p^g)\). Left: \(T=6\)h, right: \(T=48\)h.

Figure B.2: Integrated distributions of horizontal (left) and vertical (right) parcel displacements during a period \(T = 6, 12, 24, 48\)h.
B.1. STARTING GRID ANALYSIS

Figure B.3: Time variability of the total exchange amount of each instantaneous maps for the starting grid configuration corresponding to \(dM_0\)

**Instantaneous maps**

Having shown that varying \(dM\) does not introduce a bias on the overall exchange amount, estimates on the individual time intervals are now inspected. Here, we use the term "instantaneous maps" to designate the maps representing the geographical distribution of exchanges estimated with the considered \(T\).

Figure B.3 represents the time evolution of the total exchanged elementary mass volumes of each instantaneous maps with the starting grid corresponding to \(dM_0\). The four curves, corresponding to four different values of \(T\), show a satisfying consistency. For example, values with \(T=12\)h are close to the averaged values of the \(T=6\)h curve. The temporal evolution exhibits large variations, which can’t be captured with \(T \geq 24\)h anymore. This characteristic can be more precisely assessed by computing the temporal variogram (see for instance Isaaks and Srivastava 1989), which represents for a particular value of \(T\) the mean squares of the differences between values taken at the same geographical position and at two different times separated by \(\Delta t\), normalised with the variance of the entire time series. Such a variogram, which is represented in Fig. B.4 for \(T=6\)h, shows that two maps separated by \(\Delta t=6\)h have a difference which corresponds to about 60% of the statistical variability. Maps which are further separated in time have differences of the order of the statistical variability. Thus, it seems that \(T=12\)h is the maximum limit if information about the temporal variability should not be washed out.

As a further step, it is illuminating to note that the spectrum of the values of an instantaneous map is discrete due to the finite nature of the elementary volumes crossing the

---

\(^3\)Examples of such instantaneous maps can be found in Chapter 4, Figs. 4.1, 4.2 for the significant exchange events.
Figure B.4: Relative variogram of the time variability of the total exchange, for $T=6h$. A value of 1 indicates the statistical variance.

tropopause. Figure B.5 shows two examples of histograms for two different sizes of elementary mass volumes: $(\Delta x^g = 0.25^\circ, \Delta p^g = 5\text{hPa})$ and $(\Delta x^g = 0.5^\circ, \Delta p^g = 10\text{hPa})$; for a resolution of the instantaneous map $1^\circ \times 1^\circ$. The histograms have been averaged for all available instantaneous maps. Note that for one curve, the spectrum peaks are smoothed due to the finite bins of the histogram. Thus, although these two elementary mass volumes are both reasonably sized, their spectrum resolutions are very different.

The space between spectrum peaks is obviously determined by the elementary mass $dM$. On the other hand, it is straightforward that reducing the resolution of the geographical maps of exchange estimates by increasing the elementary areas $\Delta A$ has the consequence to increase the number of exchange events on each elementary area. Thus, reducing the resolution of instantaneous maps induces the stretching of the related spectrum, and in particular of the variance of its values. This led us to define a spectrum resolution $R$ by the number of peaks in the region of the spectrum delimited by twice the standard deviation of the values:

$$R = \frac{2\sigma}{dM}, \quad \text{where} \quad \sigma = \sqrt{\text{variance}} \quad (B.1)$$

It follows that the spectrum resolution can be improved either by increasing $\sigma$ via a reduction of the resolution of the instantaneous maps, or by reducing $dM$ through an increase of the number of computed trajectories.

A too low spectrum resolution is likely to lead to a degenerated geographical map, where each pixel can only realize a few number of values, resulting in patchy patterns which are not appropriate for the physical interpretation and statistical comparison with other maps. Values of $R$ larger than $R_{\text{crit}} \approx 10$ are supposed to give satisfactory results.\footnote{This critical value corresponds to an intermediate configuration compared to the spectrums shown in...} To prevent
negative effects related to a too low spectrum resolution, the configurations with low values of $R$ must be avoided. And therefore $R$ must be estimated in all our configurations, that is, for each combination of our starting grid parameters $(\Delta x^g = \Delta y^g, \Delta p^g)$, time intervals $T$, and geographical map resolutions $(\Delta x^{gm} = \Delta y^{gm})$.

The evaluation of $R$ is based on the evaluation of $\sigma$. This latter has been calculated for each configuration using second moment estimator for $\sigma$:

$$\sigma \approx \hat{\sigma} = \sqrt{\frac{1}{N-1} \sum_{i=1}^{N} (m_i - \bar{m})^2}, \quad (B.2)$$

where the $m_i$ is the value of the pixel $i$ in the concerned instantaneous map.

An other way to compute these standard deviations would be to use the similarity of this problem of arrival of elementary volumes on an elementary area during a time interval with the Poisson process. But the large discrepancies between theoretical Poisson distributions (not shown) and associated experimental distributions indicate that basic assumptions of the Poisson process are not satisfied here, and as a consequence, the Poisson $\sigma$ estimator is not appropriate. This disagreement points out that inhomogeneities in the events incidence and/or the statistical dependency of an event with an other are not negligible in the statistics of the problem.

Beside the already discussed variation of $\sigma$ with the geographical map resolution and $T$, $\sigma$ can be influenced by various other processes, like for example, the absolute number of

Fig. B.5: the peak-wise histogram represent a spectrum with $R \approx 5$, i.e. about five peaks in the main part of the spectrum, and the smooth curve $R \approx 25$. 

![Image: Histogram of exchanged mass, normalized to dM0 [au].]
events in each areas of the geographical maps, or the resolution of the spectrum itself. For
the purpose here which is to practically indicate whether a configuration allows a sufficient
spectrum resolution, it is very useful to reduce the dimensionality of $\sigma$ to its dominant
parts. To this end, a linear weighted regression $\tilde{\sigma}$ of $\dot{\sigma}$ has been performed with the expected
dominant variables: the elementary "volume" of the instantaneous map $(\Delta A \, T)^5$ and the
elementary mass of the event $(\Delta M)^6$. The resulting determination coefficient of the regression
was 0.96, which *a posteriori* justify our choice of the variables.

$$\tilde{\sigma}(\Delta x^{sg}, \Delta y^{sg}, \Delta p^{sg}; \Delta x^{sm}, \Delta y^{sm}, T) \approx \tilde{\sigma}(\Delta M, \Delta A \, T) = a \, \Delta M + b \, \Delta A \, T + c \quad (B.3)$$

Figure B.6 represents the values of $R$ as a function of $\Delta M$ and $\Delta A \, T$, calculated via
(B.2) and using the regression $\tilde{\sigma}$ as estimation of $\sigma$. This picture shows the condition
$R > 10$ strongly limits the possible values for $\Delta M$ and $\Delta A \, T$, and puts light on the config-
urations leading to degenerated maps. For example, to be able to represent geographically
our estimates with a resolution of $1^\circ \times 1^\circ$ and $T=12h$ (corresponding to $2 \, \Delta A \, T / (\Delta A_0 \, T_0)$
on the y-axis of Fig. B.6), we have to compute elementary volumes smaller then $0.5^\circ \times 0.5^\circ$
$\times 10$hPa (8 $\, \Delta M / \Delta M_0$ on the x-axis of Fig. B.6), which corresponds in our case study to the
computation of $10^6$ trajectories for every time interval of 12h. An instantaneous map with a
resolution of $3^\circ \times 3^\circ$ and $T=48$h would correspond to a value of $72 \, \Delta A \, T / (\Delta A_0 \, T_0)$ on the
y-axis of Fig. B.6, and an elementary mass $\Delta M = 3^\circ \times 3^\circ \times 30$hPa would lead to a value of
864 $\, \Delta M / \Delta M_0$ on the x-axis.

**Spatial structures**

The previous criterion concerning the spectrum resolution $R$ allows to determine the largest
elementary mass $\Delta M$ which is appropriate for a particular choice of the geographical map
resolution, in order to avoid a degeneration of the maps due to a too low spectrum resolution.
It is important to eliminate such degeneration in the geographical maps of exchange for
reasons related to the quality of the maps, but also to permit the statistical comparison of
these maps via for instance linear correlations. Calculating correlations between degenerated
instantaneous maps would lead to misleading coefficients.

Thus, under the condition that $R > 10$, correlation coefficients between any instant-
aneous map and the map calculated with the smallest elementary mass $\Delta M_0$ provide a
reasonable measure of agreement of the spatio-temporal structures of estimated exchange.
These correlations permit to verify the general "convergence" of the maps towards the map
calculated with the smallest elementary masses $\Delta M_0$, and also to analyse the sensitivity of
spatial(-temporal) structures to the horizontal and vertical components of the elementary
mass volumes $\Delta x^{sg} \times \Delta y^{sg}$ and $\Delta p^{sg}$. The justification for taking a linear correlation esti-
imator comes from the scatter plot analysis (not shown) which shows remarkably linear
relationship. Hereafter, the resolution of the geographical map of exchange is chosen to
$\Delta A_0$: $1^\circ \times 1^\circ$.

---

\(^5\)The roles of $\Delta A$ and $T$ are similar for the spectrum of the values of instantaneous maps: an increase of
either the pixel's size $\Delta A$ or the integration time interval $T$ leads to a larger number of events on each pixel of
the instantaneous maps.

\(^6\)The role of the elementary mass $\Delta M$ for the spectrum of the values of instantaneous maps is related to
the better geographical distribution of the events for larger number of events.
Figure B.6: Estimated values of $R$ (iso-lines) using the regression of $\sigma$ as a function of $dM / dM_0$ (x-axis) and $\Delta A T / (\Delta A_0 T_0)$ (y-axis). $\Delta A_0 = 1° \times 1°$, $T_0 = 6h$, $dM_0 = 0.25° \times 0.25° \times 5hPa$. An instantaneous map with a resolution of $3° \times 3°$ and $T=48h$ would correspond to a value of $72 \Delta A T / (\Delta A_0 T_0)$ on the y-axis of Fig. B.6; and an elementary mass $dM = 3° \times 3° \times 30hPa$ would lead to a value of $864 dM / dM_0$ on the x-axis.

Figure B.7 represents these correlations (shaded) for $T=6h$ and $T=48h$. Lines of constant elementary mass $dM$ are over-plotted in a range between zero and the largest elementary mass for which $R > 10$. The square grids intersections represent the computed points.

These correlation patterns suggest that most of the region where $R > 10$ exhibits a correlation greater than 0.90 in both cases. They also show that for a constant mass $dM$ it is preferable to choose a fine vertical resolution.

The “convergence” of the maps with decreasing elementary volumes to a limit map which is near our map of smallest volume configuration is confirmed by these correlation patterns, in particular for the case $T=48h$ where not less than 10 independent computations have a correlation higher than 95% with the map of smallest elementary mass configuration. A further confirmation of this convergence is given by the comparison of the spatial variograms of the maps calculated with the elementary mass configurations which are within the zone of correlation $> 0.95$ of Fig. B.7. Figure B.8 shows these variograms for the same time interval $T$ as before ($T=6h$ and $T=48h$). It is apparent that for the two cases the considered maps possess spatial structures with similar characteristics.

**Sensitivity to the data resolution**

The analysis has been repeated for the range of data resolutions listed in Table 3.1. Panels B.9, B.10 and B.11 are the equivalents of Fig. B.7 for the other resolutions. The previous general results still hold for these lower resolutions, in particular the gray bold line indicating a $R = 10$ corresponds to correlation values between 0.9 and 0.95 for reasonably small values.
Figure B.7: Correlations between the instantaneous maps with a resolution of $1^\circ \times 1^\circ$ corresponding to the smallest elementary volume configuration ($0.25^\circ \times 0.25^\circ \times 5\text{hPa}$) and those corresponding to the other configurations. X-axis and y-axis are respectively the horizontal and vertical size of the elementary volume. Gray dotted lines correspond to lines of equal $dM$ in the range between zero and the largest acceptable volume (bold line) for $R > 10$. Left panel for $T=6\text{h}$, right panel for $T=48\text{h}$.

Figure B.8: Variograms of maps corresponding to the configurations found within the $> 95\%$ correlation sector of fig B.7. Left: three configurations for $T=6\text{h}$, Right: ten configurations for $T=48\text{h}$. 
of $\Delta p^g$, and these correlation values are enhanced for larger values of $T$. These panels allow the determination of the optimum starting grid for data resolutions within the range studied here.

**Summary**

To conclude, it has been shown that (i) if a temporal continuity of the exchange maps is desired, $T$ should not overpass 12h; (ii) the elementary mass $dM$ must be chosen with regard to the resolution of instantaneous maps in order to keep a reasonable spectrum resolution ($R > 10$ in Fig. B.6); (iii) the horizontal and vertical components of the elementary mass should be chosen with a small enough vertical component (Figs. B.7, B.9 and B.10). Figure B.6 also provides the relation between the computational effort (via the size of the elementary mass, i.e. the number of trajectories) and the possible resolution for the map representing the exchange estimates.

No significant sensitivity with the data resolution has been found for any part of the analysis. Note finally that this analysis shows the important constraints on the computation procedure in the context of a case study. In the case of a climatology, it is clear that the averaging procedure would wash out the sensitivities emphasised here, and the associated constraints would not be useful.
Figure B.9: Same as Fig. B.7 but for data resolutions: Top: (0.5°, 3h), Bottom: (0.5°, 6h). Left: $T=6h$, right: $T=48h$. 
Figure B.10: Same as Fig. B.7 but for data resolutions: Top: (1.0°, 1h), Bottom: (1.0°, 6h). Left: $T=6h$, right: $T=48h$. 
Figure B.11: Same as Fig. B.7 but for data resolutions: (2.0°, 1h). Left: T=6h, right: T=48h.
B.2 Critical Parameters

B.2.1 Critical Residence Time

The residence time criterion aims at discriminating spurious exchanges which are induced by numerical errors from the significant ones. Purely numerically-induced exchanges are expected to be associated with oscillations of trajectories around the tropopause, or with an abrupt PV change along the trajectories (see section 3.2).

In order to choose an appropriate critical residence time, the tropopause crossing activity has been analysed inside the EM domain. To this end, trajectories with a length of 90 hours have been calculated from a three-dimensional basic starting grid, and the time interval between two consecutive tropopause crossings has been represented for all trajectories in the form of a residence time frequency histogram (see Fig. B.12) for our range of data resolutions. The analysis has been performed for two starting dates: forward trajectories from Sept 1 00UTC, and backward from Sept 4 18UTC. These two calculations show similar results, and therefore are represented together here.

Figure B.12 shows the distribution of the absolute number of tropopause crossings followed by a specific staying time. The term staying time is used here to designate the residence after crossing, in distinction to the residence time which is used in this study for a symmetric residence before and after the crossing. The distributions represent the total number of events found in the whole set of trajectories for several data resolutions. The sensitivity to the resolution is clearly very high for small staying times and almost absent for staying times larger than about 8h. The sensitive zone exhibits itself two different responses whether the temporal or the spatial resolution are concerned. A decrease of the spatial resolution leads to an increase of the crossing activity associated with small residence times. On the other hand, the temporal resolution effect is to homogenise the domain of staying times smaller than the temporal resolution. Both responses are obvious to interpret, the first is due to the increasing inconsistency between trajectory advection and PV computation as the spatial resolution decreases, while the second is due to the linear temporal interpolation scheme used to calculate wind and PV-fields at times inbetween data times.

The fact that the absolute number of events agrees for all resolutions for staying times larger than about 8h is very delighting. It shows that the major effects of the data resolution are limited to increasing or decreasing specifically the crossing activity which is associated with very short staying times. The choice of a good critical residence time $\tau_{crit}$ is therefore rendered unambiguous, and furthermore, resulting estimates are expected to have a certain robustness with respect to the data resolution. This provides justification for the choice of the residence time based discriminator of the discussion of section 3.3.

It can be assumed that the symmetric problem, i.e. the staying time distribution before crossing event gives similar results. Selection of significant exchanges can then be done by discriminating exchanges which experience a small staying time before and after the transition period.

Finally, a value of $\tau_{crit} = 12h$ has been chosen in this study to discriminate numerically induced exchanges, accordingly to Fig. B.12.
B.2.2 Critical Transition Time

As discussed in Chapter 3, our exchange model is based on the idea that trajectories can possibly experience multi-crossings during their transition period across the tropopause. The transition time period lies inbetween the two residence time periods occurring before and after the exchange (see Fig. 3.2). This transition time period has to be smaller than the critical residence time in order to avoid confusion between the residence period (which is regarded as the chemical origin/destination) and the transition period.

Figure B.13 represents the relative number of exchanges, selected with several values of \( \tau_{\text{crit}} \), corresponding to a choice of critical transition time \( T_{\text{trans}}^{\text{crit}} \), again for our range of data resolutions. The sensitivity to data resolution is rather weak. A temporal resolution decrease induces obviously a truncation at transition times comparable to the temporal resolution, while the spatial resolution do not have a clear overall influence. On the other hand, these distributions are quite sensitive to the choice of \( \tau_{\text{crit}} \), but in an expected way: small critical residence times exclude slow oscillations in the transition, and therefore tend to favour small transition time periods.

Focussing on the highest resolution, represented on panel (a) of Fig. B.13, it is found that a choice of \( T_{\text{trans}}^{\text{crit}} \approx \tau_{\text{crit}} \) removes 20% of exchange events, and that a \( T_{\text{trans}}^{\text{crit}} \) half as large as \( \tau_{\text{crit}} \) removes about 30%. These ratios are smaller for other data resolutions. In this study, the transition time is everywhere taken as \( T_{\text{trans}}^{\text{crit}} = 1/2 \tau_{\text{crit}} \) to ensure a larger residence time period in the stratosphere/troposphere than in the region of the tropopause.
Figure B.13: Relative number [%] of exchanges with a transition time smaller than the specified transition time, for several values of the critical residence time $\tau_{\text{crit}}$. Data resolutions: (a) 0.5° 1h, (b) 0.5° 3h, (c) 0.5° 6h, (d) 1.0° 1h, (e) 1.0° 6h, (f) 2.0° 1h.
B.3 Effects of the Resolution Transition

The resolution of global data has been enhanced regionally via the mesoscale EM model. This permits to improve the accuracy of trajectory computation and preliminary exchange selection inside the model domain (see Chapter 3). However, according to our exchange model, trajectories must be prolonged for the selection of significant exchange events via the residence time criterion. The prolonged trajectories may cross EM domain boundaries and therefore experience a change in the underlying data resolution. This resolution change together with possible effects related to the relaxation boundary of the model could induce a bias in the residence time selection. It is therefore necessary to analyse the effect of crossing the EM domain boundary on the trajectory residence time.

The trajectory residence time selection is influenced through the probability that a trajectory still residing in either the troposphere/stratosphere crosses the 2PVU surface for the first time. The latter probability will be called crossing probability in the following. Two kind of effects can be expected, namely (a) direct effects of the boundary of the model domain on the crossing probability of trajectories, and (b) bias induced by a possibly different crossing probability outside the EM domain.

B.3.1 Effect of EM Domain Boundary

Effects of the model boundary can be emphasised by comparing the crossing probability experienced by a trajectory while going across the boundary, with the crossing probability experienced before having crossed the boundary. However, this case study does not allow a comparison which is independent of the particular synoptic evolution, and differences are due to both the synoptic evolution and the resolution transition. Here, two different sets of 90 hours trajectories, for two different synoptic evolutions, have been used in concert and analysed together with an eye on the synoptic evolution. Figure B.14 represents the crossing probability inside the domain and at the transition zone (i.e. EM boundary ± 5°). The first set shows an increased crossing activity in the boundary region after 40h, which is not visible inside the domain. Looking at the synoptic evolution, a positive PV anomaly is found located on the south eastern boundary of the EM domain for the whole time period 40h - 80h. This anomaly partly dilutes and decays. A remnant of this anomaly crosses the diagnosis boundary at Sept 4 12UTC and causes an intense upward exchange zone. The difference found in the two curves in panel (a) can therefore be attributed to this PV destruction in the south east region of the EM domain boundary. However, the possibility for unrealistic tropopause crossings due to the boundary relaxation which does not necessarily conserve PV can not be neglected.

The second set of trajectories whose crossing probabilities are given in Fig. B.14 (b) shows a similar crossing probability inside the domain and at its boundaries for the period up till 60h. The strong increase inside at about 70h is due to a large downward exchange event attributed to the rapid erosion of the streamer. This event entirely develops inside the domain and is therefore not seen in the boundary region.

This discussion shows that the possibility for unrealistic tropopause crossings associated with synoptic events near the model boundary can not be completely eliminated. This kind
B.3. EFFECTS OF THE RESOLUTION

Figure B.14: Crossing probability of trajectories with length of 90 hours started (a) forward on the 1st Sept. 00UTC, and (b) backward on the 4th Sept. 18UTC. Only trajectories which cross the 2PVU surface are taken into account. Solid line: crossing activity inside the EM domain, dashed line: crossing activity in the boundary belt (=± 5°). Trajectories have been computed with data resolution (0.5°, 1h).

of events can enhance artificially the crossing probability and distort the selection related to the residence time. In the case of the first set of trajectories, and assuming that all crossings that occurred after 40h within the boundary region were unrealistic, the part of erroneous crossings to the total is 5.5%. Thus, even if all crossings occurring in the boundary region were unrealistic, their overall importance remains low.

B.3.2 Sensitivity to the Resolution

A possible bias in the residence time selection due to a change in the crossing probability associated with a change in the data resolution can be assessed by looking at the crossing probabilities for data resolutions of (0.5°, 1h) and (1.0°, 6h) inside the EM domain. The crossing probabilities can be compared directly because they represent the same synoptic evolution. Figure B.15 indicates that results are very similar for the two sets of trajectories. The total number of crossings differs by 12% in the first set and by 2% in the second.

This points out the fair robustness of the residence time selection to the resolution change, and demonstrate the reliability of this criterion for the exchange quantification in the context of a case study.
Figure B.15: Crossing probability of 90h trajectories started (a) forward on the 1st Sept. 00UTC, and (b) backward on the 4th Sept. 18UTC. Only trajectories which cross the 2PVU surface are taken into account. Solid line: crossing activity with data resolution (0.5°, 1h), dashed line: (0.5°, 1h). Crossing events have been selected to occur inside the EM domain.
Table B.3: Correlation coefficients between geographical distributions computed using incoming and outgoing points as exchange locations, for various data resolutions. Values are provided for the time mean and time minima of correlations.

### B.4 Sensitivity of STE Maps with the Exchange Location Convention

The exchange model introduced in 3.2 has been proposed with a convention upon the exchange location along the trajectory. The exchange location has been conventionally placed at the “incoming point”, i.e. at the end of the transition period. The decision of using this point rather than an “outgoing point”, similarly defined as the first point of the transition period, comprises some arbitrariness and it is important to quantify the sensitivity of the geographical exchange maps to this definition.

To this end, additional geographical distributions of STE have been computed using the outgoing point definition for a range of data resolutions, and compared to geographical distributions of STE computed with the conventional incoming point.

The comparison of geographical distribution (not shown) shows a very weak sensitivity. A quantification of this sensitivity is provided in table B.3 via the correlation between geographical distributions. Time means and time minima are given for stratosphere-to-troposphere and troposphere-to-stratosphere fluxes, respectively. Mean correlations are always larger than 0.94, while the smallest correlation found at any time period and data resolution is 0.89. The correlation is improved when the time resolution is lowered, while it decreases when the spatial resolution is weakened. This sensitivity is directly related to the gap between the two exchange possible points, which is itself influenced by the oscillations caused by numerical errors and inconsistencies. The behaviour is therefore in agreement with the fact that such oscillations are favoured at low spatial resolution and disfavoured at low temporal resolution, as discussed in Appendix B.2.

Finally, the very high correlations permit to neglect this sensitivity and to focus exclusively on the incoming point convention to determine the exchange location.
Appendix C

Sensitivity of Estimated STE to the Data Resolution in a Case Study

The proposed method has been constructed in Chapter 3 and various analyses (dimensions of the starting grid, critical residence time, critical transition time, change of data resolution at the EM domain boundary) have been conducted to check and optimise its reliability a priori. In Chapter 4 it has been shown that the results are consistent with the structure of the tropopause and the results were qualitatively validated on the mesoscale. Total exchange fluxes are comparable to typical values found in the literature. In this Appendix, the response of the method to the data resolution is discussed in order to check the reliability of the method a posteriori and the possibility to apply it to lower resolutions.

The sensitivity of exchange estimates to the data resolution is related to two factors. One is the scale of the physical and dynamical processes that lead to exchange. Data resolution and requisite calculations (computation of derivatives and interpolations) imply the truncation of small scales in the scale spectrum and consequently exclude the corresponding STE events. The tropopause is smoothed by the potential vorticity calculation and is sensitive to the spatial data resolution. The exchange flux computation on the other hand, is sensitive to both spatial and temporal resolution via the trajectory calculation.

The second factor is numerical errors due to finite data resolution. One effect of this kind is the lack of coherency between the computed trajectories and the tropopause (section 3.2), and is in effect limited by applying the residence time criterion. Another effect is the numerical deviation of trajectories from the “true” path caused by the sparse temporal data resolution. Doty and Perkey (1993) showed that in the case of an intense extratropical cyclone, trajectory computations can be sensitive to the temporal data resolution. As here, they used data degraded in time, originally generated using a mesoscale model, to advect trajectories following the Petterssen method (see section 3.2). They showed that the deviation of trajectories computed with low (degraded) temporal resolution from trajectories computed with data at every model time step (15 min) become significant for data time steps of 3 h and longer.

Table C.1 lists the mean STE fluxes for our range of data resolutions. Sensitivity is found for both temporal and spatial data resolution. A lowering of the temporal resolution implicates an increase in exchange estimates in both directions which almost cancel in the
APPENDIX C. SENSITIVITY TO THE DATA RESOLUTION

<table>
<thead>
<tr>
<th>Resolution</th>
<th>S $\to$ T</th>
<th>T $\to$ S</th>
<th>NET (S$\to$T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5°, 40 lev, 1 h</td>
<td>2.2</td>
<td>1.6</td>
<td>0.6</td>
</tr>
<tr>
<td>0.5°, 40 lev, 3 h</td>
<td>2.4</td>
<td>2.0</td>
<td>0.4</td>
</tr>
<tr>
<td>0.5°, 40 lev, 6 h</td>
<td>3.0</td>
<td>2.6</td>
<td>0.4</td>
</tr>
<tr>
<td>1.0°, 31 lev, 1 h</td>
<td>2.2</td>
<td>2.1</td>
<td>0.1</td>
</tr>
<tr>
<td>1.0°, 31 lev, 6 h</td>
<td>2.8</td>
<td>2.9</td>
<td>-0.1</td>
</tr>
<tr>
<td>2.0°, 31 lev, 1 h</td>
<td>1.9</td>
<td>1.9</td>
<td>0.0</td>
</tr>
</tbody>
</table>

Table C.1: Temporally and spatially integrated exchange mass fluxes for the range of data resolutions. Unit: $10^9$ kg s$^{-1}$. Integration domain: 10W-20E, 38N-58N.

The spatial resolution effect is less marked and preferentially affects the troposphere-to-stratosphere exchange direction.

The temporal evolution of the exchange estimates for various data resolutions is shown in Fig. C.1. The stratosphere-to-troposphere exchange has been shown (section 4.1) to occur predominantly within distinct zones associated with the streamer ($t \sim 0 - 30$ h) and cut-off ($t \sim 30 - 80$ h). Here, the discussion focusses on the stratosphere-to-troposphere fluxes, and sensitivities are interpreted separately for the major exchange episodes and for the remaining, namely the background.

The sensitivity to spatial resolution leads clearly to distinct features. The changes between 0.5° and 1° are weak and occur only during the cut-off decay phase. Inspecting the tropopause structure with the 1° resolution shows that the cut-off remains an entity, without cut isolated blob, as an effect of the lowered vertical resolution (31 levels). Furthermore, the lowest part of the cut-off is smaller in diameter and possesses smaller PV values than with the 0.5° data resolution. The exchange processes identified with the 0.5° resolution have small-scales which are therefore not properly resolved with resolutions lower than 1°. Consequently, it is not a surprise to see at the 2° resolution a further reduction of the flux in the latest phase of the cut-off decay. On the other hand, the streamer break-up episode, which is still very well reproduced at 1°, is strongly underestimated with the lowest resolution (2°). Finally, the “background” exchange activity, which actually takes place within distinct secondary patches with spatial extents of hundreds of kilometres, is favoured on the average when the spatial resolution is lowered. This effect is also present in the troposphere-to-stratosphere exchange, in particular with the 1.0° resolution.

The sensitivity to the temporal data resolution, on the other hand, does not exhibit such a dependency on the specific phase of the evolution (Fig. C.1). The 1 h and 3 h resolutions give very close estimates, both in the total and the geographical distribution. An exception occurs however for the period 12 - 24UTC of the 3rd. Geographical distributions show that the difference is due purely to an exchange event over Scotland which is strengthened in the 3h resolution. The event is characterised by large vorticity values on the tropopause ($> 3.5 \times 10^{-4}$ s$^{-1}$), even exceeding values within the cut-off. The time evolution of exchange flux estimates using the 6h resolution compares well with the one at 3h, but with a systematic shift towards larger values. The comparison of the geographical distributions shows that while the location and intensity of the major events are similarly reproduced, some secondary patches of exchange are reinforced when using the 6h data resolution. Thus, the inaccuracies in trajectory computation caused by temporally sparse data highlighted by Doty and Perkey...
Figure C.1: Time evolution of the horizontally integrated mass exchange, estimated with several data resolutions and $\tau_{\text{crit}} = 12$ h. Left: stratosphere-to-troposphere exchange, right: troposphere-to-stratosphere exchange. Time relative to the 1st of September at 00UTC. Values are drawn at the centre of 12 h integration periods. Unit: $10^9 \text{kg s}^{-1}$. Integration domain: 10W-20E, 38N-58N.

(1993) take place here in individual favourable events.

It is important to understand more precisely the cause for such reinforcements, because according to Fig. C.1, they play a significant role in the total flux when computed from data with 6 h temporal resolution. To further investigate this issue, exchanges which correspond to trajectories passing through regions where the flow field is expected to decrease their accuracy have been geographically represented and compared in intensity and location with the basic exchange distributions. Sensitive meteorological situations have been defined with two indicators: large vertical relative vorticity $\zeta$ and large vertical gradient of vertical velocity $|\partial w/\partial z|$. According to Seibert (1993) these two criteria are critical for the convergence and accuracy of trajectories. An additional interest in looking at $\partial w/\partial z$ stems from its appearance in the diagnostic determination of the vertical velocity:

$$\nabla_h u + \frac{1}{\rho_0} \frac{\partial}{\partial z} (\rho_0 w) = 0$$  \hspace{1cm} (C.1)
Figure C.2: Stratosphere-to-troposphere exchange mass fluxes for the periods (a) Sept 1 00-12UTC, (b) Sept 2 12-24UTC, (c) Sept 3 12-24UTC, in 10^8 kg s^-1. The first three columns from the left represent respectively, exchanges associated with $\zeta > 2 \times 10^{-4}$ s^-1, exchanges associated with $|\partial w/\partial z| > 0.3 \times 10^{-4}$ s^-1 and all exchanges computed with 6h data time resolution. The last column represents the reference, as computed with 1h data time resolution. Spatial resolution of the data is 0.5° everywhere. Bold lines represent the 325 and 330 K potential temperature contours at the 2PVU level at the intermediate time of each period.

In this work, the convergence of the trajectory time step computation is expected in most cases according to the choices of a small trajectory time step (1/12 of the data time step) and of the number of three iterations (see section 3.2). However, it can occur that in some extreme conditions the convergence is not reached. Such cases are associated with very low trajectory accuracy. The threshold values for $\zeta$ and $|\partial w/\partial z|$ have been chosen such that the accepted values represent less than 10% of the corresponding distribution: $\zeta_{\text{thres}} = 2 \times 10^{-4}$ s^-1 and $(\partial w/\partial z)_{\text{thres}} = 0.3 \times 10^{-4}$ s^-1. The separation of both indicators allows to enlight if the possible trajectory position error is due rather to horizontal or vertical velocities.

Figure C.2 shows a comparison of the geographical distributions of these selected exchange events with the basic ones for three integration periods. The two first columns

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1It will be interesting to further investigate this point by allowing a large number of iterations (about 20 - 40) and by verifying the convergence with a criterion based on the distance between two consecutive iterations.
Figure C.3: Time evolution of the exchanged mass estimated with several data resolutions and $\tau_{\text{crit}} = 12\text{h}$. Left: stratosphere-to-troposphere exchange, right: troposphere-to-stratosphere exchange. Time relative to Sept 1 00UTC. Values are drawn at the centre of the 12h integration periods. Unit: $10^9\text{kgs}^{-1}$. Integration domain: 10W-20E, 38N-58N.

represent exchange events computed with (0.5°, 6h) data resolution associated with a large vorticity and a large vertical gradient of vertical velocity, respectively. The third column represents the total fluxes at the same resolution, and the forth gives as reference the total fluxes computed with the highest data resolution (0.5°, 1h). The differences which can be seen between estimates with 1h and 6h data resolutions are clearly associated with strong values of at least one of the two indicators. It can therefore be concluded that the differences in exchange fluxes between computations using data resolutions at 1h and 6h are due to large values of rotation and/or horizontal divergence, which favours inaccuracies in the trajectory calculations.

Note that the combination of low temporal resolution with the fine-scale wind structures in the 0.5° resolution data set is particularly favourable for the production of such inaccuracies. This points to the importance of the consistency between spatial and temporal data resolution used for trajectory computations. In the present case, the results are not significantly better with the (1°, 6h) data set than with the (0.5°, 6h), see Fig. C.3. This is due to the particular scheme of spatial degradation used in this study. Data at 1.0° resolution is obtained by a bilinear interpolation of the basic data (0.5°), which is exactly the same, in this particular case, as picking up every second value of the basic grid. This scheme has no spatial filtering effect and even induces some physical inconsistency between the vertical velocity gradient and the horizontal divergence (cf (C.1)).

Conversely, ECMWF analysis data from the T213L31 ($\sim$ 1.0°, 6h) has spatial structures which are consistent with the temporal resolution. Furthermore, the horizontal wind divergence is reduced through the normal-mode initialisation procedure which serves to obtain (almost) balanced initial conditions for the ECMWF model integration. This procedure, which can be seen as a reduction of the vertical gradient of the vertical velocity implies a reduction of the problem of inaccurate trajectory computation, as a direct consequence of the previous discussion. The accuracy of STE estimates based upon wind field from ECMWF
analysis data is therefore expected to be significantly better than the estimates with (1.0°, 6 h) resolution in this case study. A quantitative analysis of the accuracy of STE estimates using ECMWF data is however difficult to realize because of the lack of comparable estimates based on higher resolution data. Also, a comparison between our estimates based on the model simulation with estimates based on ECMWF data would exhibit differences which rely for a significant or even dominant part on the differing synoptic development, that is on the prognostic error of the mesoscale model simulation.

To summarise this discussion, data resolution has been shown to induce different responses on STE estimates for the time and space dimensions. Sensitivity to spatial resolution is due principally to the truncation of a part of the scale spectrum of physical processes. Data with a resolution of 1.0° has properly caught the break-up of the streamer and the beginning of the cut-off decay, but not with the same accuracy the subtle decay phase of the cut-off. The amount of spatially scattered exchange events increases when the spatial resolution is decreased. This effect is thought to be mostly associated with inconsistencies in the applied degradation procedure of the three wind field components, but no explicit tests have been conducted to verify this hypothesis. The temporal resolution has weak effects in the range of 1 - 3 h. The analysis of the estimates based on (0.5°, 1 h) and (0.5°, 6 h) resolutions has shown that the differences are associated with small-scale structures in the wind field, which are themselves strongly favoured by the data degradation method used in this study. At similar temporal resolution, ECMWF reanalysis data can be expected to yield much better estimates of STE.
Appendix D

A Simple Formulation for the Chemical Forcing due to Exchange

As noticed in Chapter 5, the residence time that exchanged parcels are likely to remain within the new layer (troposphere or stratosphere) follows a statistical distribution which can be written as

\[ P_{\text{dyn}}(t) = P_{\text{dyn}}^{\infty} + (1 - P_{\text{dyn}}^{\infty})e^{-Rt}, \]  

assuming a normalisation such that \( P_{\text{dyn}}(t = 0) = 1 \). \( R \) is the rate at which parcels return in the originating layer. \( R \) can be written in terms of the characteristic residence time \( \tau_{1/2}^{\text{res}} \) via

\[ R = \ln 2 / \tau_{1/2}^{\text{res}} \]

On the other hand, exchanged parcels enter in chemical reaction with the components present within the new surrounding as soon as they have penetrated the layer. Let's denote the exchanged constituent as \( A \). Assume a two-components chemical reaction \((D.3)\), with the reactant present in the new surrounding being named by \( B \) and the product by \( C \).

\[ A + B \rightarrow C \]  

The rate of production of the constituent \( C \) is obviously a combination of the characteristic residence time and the reaction rate, and represents in a certain sense the chemical effect of the presence of the constituent \( A \).

In the following, a formulation of the chemical production rate of the species \( C \) via \((D.3)\) is proposed as indicator of the chemical forcing due to the stratosphere-troposphere exchange. The resulting formulae is applied in Chapter 9 to the one-year climatology estimates to improve the understanding of the chemical influence of STE.

D.1 Definition of the Problem

The problem is idealised in the way illustrated in fig. D.1. Tropospheric constituent \( A \) is injected within the stratosphere with a flux \( F_A \) assumed constant in space and time. Once in the stratosphere, \( A \) can either return into the troposphere, either react with \( B \) to produce \( C \).
Figure D.1: Schematic illustrating the idealised model for the formulation of the production rate of $C$. See text.

In turn, $C$ can either be injected into the troposphere, or stay inside the stratosphere. The mass of $C$ remaining in the stratosphere is the magnitude of interest here, and defines the indicator of the chemical forcing due to the troposphere-to-stratosphere flux $F_A$. Residence time distribution is assumed of the type given by (D.1) with the characteristic time $\tau_{1/2}^{res}$. $P^{dyn}(t^*)$ can be interpreted as being the probability that a parcel injected at time $t = 0$ has to be still residing in the stratosphere at a later time $t = t^*$. The considered probe chemical reaction is of the type given in (D.3) where the component $B$ is assumed to be abundant in the stratosphere whereas the component $A$ comes from the troposphere. Suppose furthermore the reaction to be a first order reaction taking place within the stratosphere. The kinetics of $A$ is then described by the linear differential equation

$$\frac{D[A](t)}{Dt} = -K_{AB}[A](t)[B] \quad (D.4)$$

$$\pm -K_A[A](t) \quad (D.5)$$

$K_{AB}$ is the reaction rate of (D.3) and is assumed constant for matter of simplicity, and therefore $K_A = K_{AB}[B] = const$. The square brackets represent here the mass concentrations. Note that the latter hypothesis implies the independence of the chemical reaction rate with temperature, pressure and radiation. More usefully here, the kinetic equation can be written in term of the masses found in a control volume

$$\frac{Dm_A(t)}{Dt} = -K_A m_A(t) \quad (D.6)$$

and similarly, for the production of $C$

$$\frac{Dm_C(t)}{Dt} = K_A m_A(t) \quad (D.7)$$

The solution of (D.6) is obviously the exponential

$$\frac{m_A(t)}{m_A0} = e^{-Ka}$$

$$ (D.8)$$
and the chemical lifetime of $A$ is given by definition via

$$\tau_{1/2}^{\text{chem}} = \ln 2 / K_A \quad (D.9)$$

### D.2 Decay of a Pulse Release

Consider a pulse of troposphere-to-stratosphere exchange released at time $t_1$ (see picture D.1) across a surface element $dS$ of the tropopause. The mass of $A$ injected into the stratosphere during the pulse (lasting $dt$) is in general $dm_A(x, y, z, t) = F_{A}^{T \rightarrow S} \cdot dS dt$. For simplicity, we assume a horizontal tropopause without multi-tropopause structures (i.e. $dS = dx dy$) and a flux $F_{A}^{T \rightarrow S}$ constant in time and space. The pulse delivers then a mass

$$dm_A = F_{A}^{T \rightarrow S} dx dy dt \quad (D.10)$$

of $A$ in the stratosphere. After $t_1$, the mass of $A$ decreases by two ways: (1) by chemical reaction with $B$ producing $C$, and (2) by returning to the troposphere conformly to the residence time distribution. The probability that a mass element remains in the stratosphere up till a time $t_2$ is the product of the probability $P_{A}^{\text{chem}}(t_2 - t_1)$ that the mass element has not chemically reacted during $(t_2 - t_1)$, with the probability $P_{A}^{\text{dyn}}(t_2 - t_1)$ that the mass element has resided within the stratosphere during $(t_2 - t_1)$, with

$$P_{A}^{\text{dyn}}(t) = P_{A}^{\text{dyn}} + (1 - P_{A}^{\text{dyn}}) e^{-R_AT} \quad (D.11)$$

$$P_{A}^{\text{chem}}(t) = e^{-K_AT} \quad (D.12)$$

Thus, the mass of $A$ remaining at time $t_2$ from a pulse at time $t_1$ can be written as

$$dm_{A}^{\text{rem}}(t_2; t_1) = F_{A}^{T \rightarrow S} P_{A}^{\text{dyn}}(t_2 - t_1) P_{A}^{\text{chem}}(t_2 - t_1) dx dy dt_1 \quad (D.13)$$

Let’s look in details at the mass of $C$ $dm_{C}^{\text{prod}}(t_2; t_1)$ which is produced at time $t_2$ from the remaining mass of $A$ of the pulse released at $t_1$. The pure chemical production rate of $C$ is given in (D.7), and knowing the expression (D.13) for the remaining mass of $A$, we can write

$$dm_{C}^{\text{prod}}(t_2; t_1) = K_A dm_{A}^{\text{rem}}(t_2; t_1) dt_2 \quad (D.14)$$

Finally, the mass of $C$ $dm_{C}^{\text{rem}}(t^*; t_2; t_1)$ which is still within the stratosphere at an ulterior time $t^*$ is the part of $dm_{C}^{\text{prod}}(t_2; t_1)$ which has not returned into the troposphere between $t_2$ and $t^*$.

$$dm_{C}^{\text{rem}}(t^*; t_2; t_1) = dm_{C}^{\text{prod}}(t_2; t_1) P_{C}^{\text{dyn}}(t^* - t_2) \quad (D.15)$$

### D.3 Final Expression

To estimate the accumulated production of $C$ due to the flux of tropospheric mass across an element of tropopause, it remains to integrate the expression of $dm_{C}^{\text{rem}}(t^*; t_2; t_1)$ for $t_2 \in [t_1, t^*]$
and for \( t_1 \in [0, t^*] \).

\[
M_c(t^*) = \iint_{t_1 \in [t_1, t^*]} dm_c^{em}(t^*; t_2; t_1)
\]

\[
= dx dy K_A P_A^{T \to S} \iint_{t_1 \in [t_1, t^*]} dt_1 dt_2
\]

\[
P_A^{dyn}(t_2 - t_1) P_C^{dyn}(t^* - t_2) P_A^{chem}(t_2 - t_1)
\]

The final comprehensive expression is the following:

\[
M_c(t^*) = dx dy K_A P_A^{T \to S} \left\{ \frac{1}{K_A} \left( \frac{P_{A\infty} P_{C\infty}}{K_A} - \frac{P_{A\infty} (1 - P_{C\infty})}{R_C - K_A} \right) e^{-K_A t^*} \right. \\
- \frac{(1 - P_{A\infty}) (1 - P_{C\infty})}{(R_C - R_A) R_A} e^{-R_A t^*} \\
+ \frac{(1 - P_{A\infty}) P_{C\infty}}{(R_A + K_A)^2} e^{-(R_A + K_A) t^*} \\
+ \frac{1}{R_A} \left( \frac{P_{A\infty} (1 - P_{C\infty})}{R_C - K_A} + \frac{(1 - P_{A\infty}) (1 - P_{C\infty})}{R_C - R_A} \right) e^{-R_A t^*} \\
+ \left( - \frac{P_{A\infty} P_{C\infty}}{K_A^2} + \frac{P_{A\infty} (1 - P_{C\infty})}{(R_C - K_A) K_A} \frac{P_{A\infty} (1 - P_{C\infty})}{R_C - R_A} \right) \\
- \frac{(1 - P_{C\infty})(1 - P_{C\infty})}{R_C - R_A} + \frac{(1 - P_{A\infty})(1 - P_{C\infty})}{R_A (R_C - R_A)} \\
- \frac{(1 - P_{A\infty}) P_{C\infty}}{(R_A + K_A)^2} \\
+ \left( P_{A\infty} P_{C\infty} \frac{1 - P_{A\infty}}{R_A + K_A} \right) t^* \}
\]

D.4 Stationary State

In the present context, we are interested in the production rate of \( C \) in the stationary limit when transient adjustments are not yet dominant, or in other words when \( t^* \) is large. The four first terms of the latter expression are cancelled for large values of \( t^* \) while the fifth stays constant. The only term expressing a production rate at large \( t^* \) is the last one.

\[
M_c^{\infty}(t^*) = \lim_{t^* \to \infty} M_c(t^*)
\]

\[
= dx dy F_A^{T \to S} P_{C\infty} \left( P_{A\infty} + \frac{1 - P_{A\infty}}{1 + \frac{R_A}{K_A}} \right) t^*
\]
lifetime $\tau_{1/2}^{\text{chem}}$, the characteristic residence time $\tau_{1/2}^{\text{res}}$ and the infinitely remaining part of the residence time distributions $P_{A\infty}$ and $P_{C\infty}$. This factor, which is called forcing efficiency of exchange $\mathcal{E}^{T\rightarrow S}$ here, can be used as a first order approximation of the chemical forcing that a material exchange between the troposphere and the stratosphere potentially induces.

\[
\mathcal{E}^{T\rightarrow S} = P_{C\infty} \left( P_{A\infty} + \frac{1 - P_{A\infty}}{1 + \frac{\tau_{1/2}^{\text{chem}}}{\tau_{1/2}^{\text{res}}}} \right)
\]  

(D.29)

In the limit of a flat residence time distribution $P_{A}^{\text{dir}}(t) = P_{A\infty} = 1$, only the first term in bracket remains in (D.29), as would be expected from the conventional conception of the exchange flux. But with the new residence properties of the exchange inferred in this thesis, a second term appears which account for the competition between characteristic residence time and chemical lifetime. Note finally that the values of $P_{C\infty}$ and $P_{A\infty}$ can be reasonably equalised by assuming that the chemical reaction has no effect on the path of the parcel.
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Bibliography


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<td>1997 - 2000</td>
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